
Mussel Watch

Recent Trends in Coastal Environmental Quality

U.S. Department of Commerce
National Oceanic and Atmospheric Administration
National Ocean Service

National Status and Trends Program

Since 1984, the National Oceanic and Atmospheric Administration (NOAA) has monitored, through its National Status and Trends (NS&T) Program, the concentrations of organic compounds and trace metals in bottom-feeding fish, shellfish, and sediments at almost 300 coastal and estuarine locations throughout the United States. The objective of the program, which is administered by the Coastal Monitoring and Bioeffects Assessment Division of the Office of Ocean Resources Conservation and Assessment, is to determine the status and long-term trends of contamination in these important areas. Samples collected annually through the program are analyzed to determine levels of synthetic chlorinated compounds (e.g., DDTs), polychlorinated biphenyls (PCBs), polynuclear aromatic hydrocarbons (PAHs), and trace metals (e.g., mercury and lead). NOAA's NS&T Program is the first to use a uniform set of techniques to measure coastal and estuarine environmental quality over relatively large space and time scales. A "specimen bank" of samples taken each year at about 10 percent of the sites is maintained at the National Institute of Standards and Technology for future, retrospective analyses. A related program of directed research is examining the relationships between contaminant exposures and indicators of biological responses in fish and shellfish (i.e., bioeffects) in areas that are shown by the NS&T monitoring results to have high levels of toxic chemicals.

This report, based on data from analyses of mollusks, describes trends in contamination from 1986 through 1990. It follows, and in some sections reiterates, a 1990 report (O'Connor, 1990) that described the spatial extent and severity of sediment contamination.

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Recent Trends in Coastal Environmental Quality: Results from the First Five Years of the NOAA Mussel Watch Project

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INTRODUCTION

Following increasing public and scientific concern about the quality of the marine environment and the absence of any long-term national monitoring program in the United States in the early 1980's, the National Oceanic and Atmospheric Administration (NOAA) created the National Status and Trends (NS&T) Program in 1984. The program monitors trends of chemical contamination and assesses the effects of human activities on coastal and estuarine areas around the Nation. It has been analyzing estuarine and coastal sediments and tissue samples from selected organisms for a broad suite of trace metals and organic chemicals. Samples are collected from a network of sites located around the coastline of the U.S. Tissues are also examined for evidence of biological response to environmental contamination such as liver tumors and reproductive damage.

Since 1986, the NOAA Mussel Watch Project, a major component of the NS&T Program, has been making the same chemical measurements on surface sediments and whole soft-parts of mussels and oysters collected from about 200 coastal and estuarine sites. Recent results from the Mussel Watch Project describe the spatial distribution of coastal contamination and, where temporal trends exist, show contamination to be decreasing in many instances. This finding implies that some benefits have resulted from the management of chemical use and discharge. However, data for more years will be necessary to distinguish the effects of human activity from those of natural influences on some of these chemical concentrations.

SAMPLING SITES AND SPECIES

The need for large-scale and long-term monitoring was emphasized by a U.S. National Research Council report (NRC, 1990) indicating that more than \$130

million is being spent every year on U.S. marine environmental monitoring, but that most of it is devoted to compliance monitoring, i.e., testing wastewaters and other materials prior to discharge, or making measurements near discharge points as prescribed by regulation. Since compliance monitoring, by design, covers very small spatial scales, national programs such as NOAA's NS&T Program are the only ones focusing on wider public concerns. It is on this wider scale that national benefits should be derived from expending billions of dollars to control direct and indirect chemical discharges to coastal and marine waters.

The Mussel Watch Project was designed to describe chemical distributions over national and regional scales. Therefore, it is important for sampling sites to be representative of large areas rather than the small-scale patches of contamination commonly referred to as "hot spots." To this end, no sites were knowingly selected near waste discharge points. Furthermore, since the Mussel Watch Project is based on analyzing indigenous mussels and oysters, a site must support a sufficient population of these mollusks to provide annual samples.

NS&T sampling sites are not uniformly distributed along the coast. Within estuaries and embayments, they average about 20 kilometers (km) apart, while along open coastlines the average separation is 70 km. Almost half of the sites were selected in waters near urban

areas, within 20 km of population centers in excess of 100,000 people. This choice was based on the assumptions that chemical contamination is higher, more likely to cause biological effects, and more spatially variable in these waters than in rural areas.

In 1986 and 1987, 145 Mussel Watch sites were sampled. In 1988, a few sites were added on the East Coast to fill in large spatial gaps between sites, and one was added in Hawaii to provide a third sampling site for an oyster species that is not sampled elsewhere. Also in 1988, 20 new sites were selected in the Gulf of Mexico for the specific purpose of gathering samples closer to urban centers.

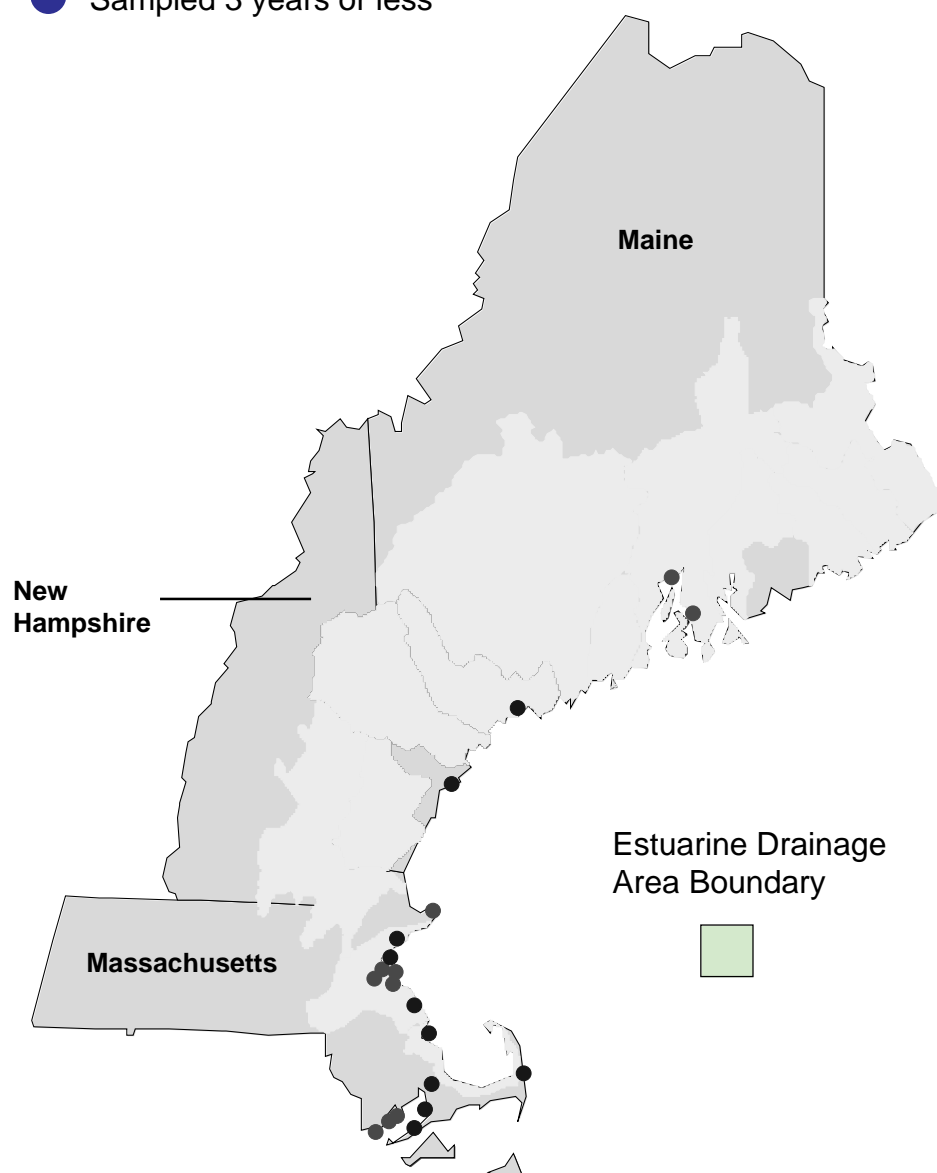
Results from the initial sampling showed that the highest chemical concentrations were near urban areas on the East and West Coasts, and that few sites in the Gulf of Mexico could be considered contaminated. Since urban centers along the Gulf are further inland than those near other coasts, an attempt was made to sample as close to them as possible. The major limitation on sampling further inland is that oysters are not found at salinities below about 10 parts per thousand. By 1990, 234 sites had been sampled, with further additions made to test the representativeness of earlier sites.

Figures 1a through 1e indicate the locations of all sites sampled in the NS&T Mussel Watch Project in the contiguous United States. Three sites are located in Hawaii, and two in Alaska. Locations of

North Atlantic Mussel Watch Sites

Figure 1a. *Mollusk collection sites 1986 through 1990.*

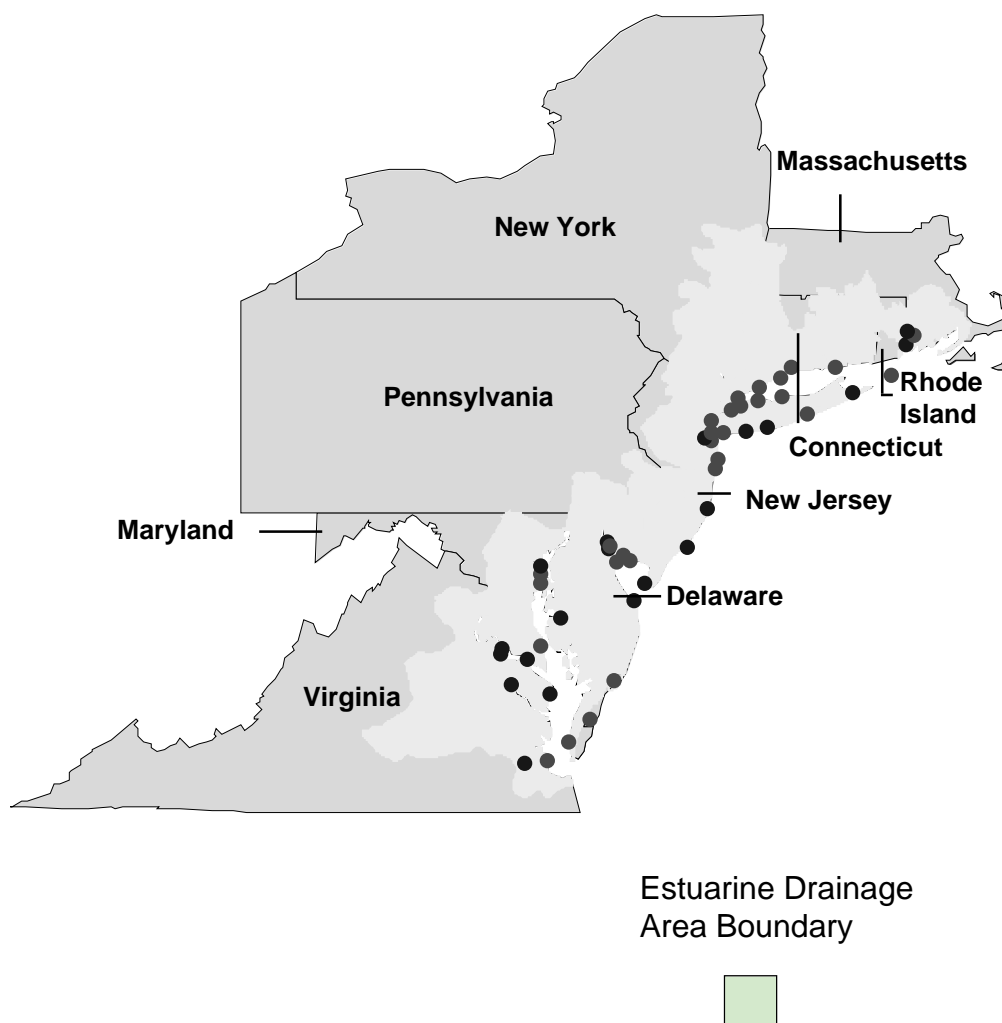
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- Sampled 3 years or less



Middle Atlantic Mussel Watch Sites

Figure 1b. *Mollusk collection sites 1986 through 1990.*

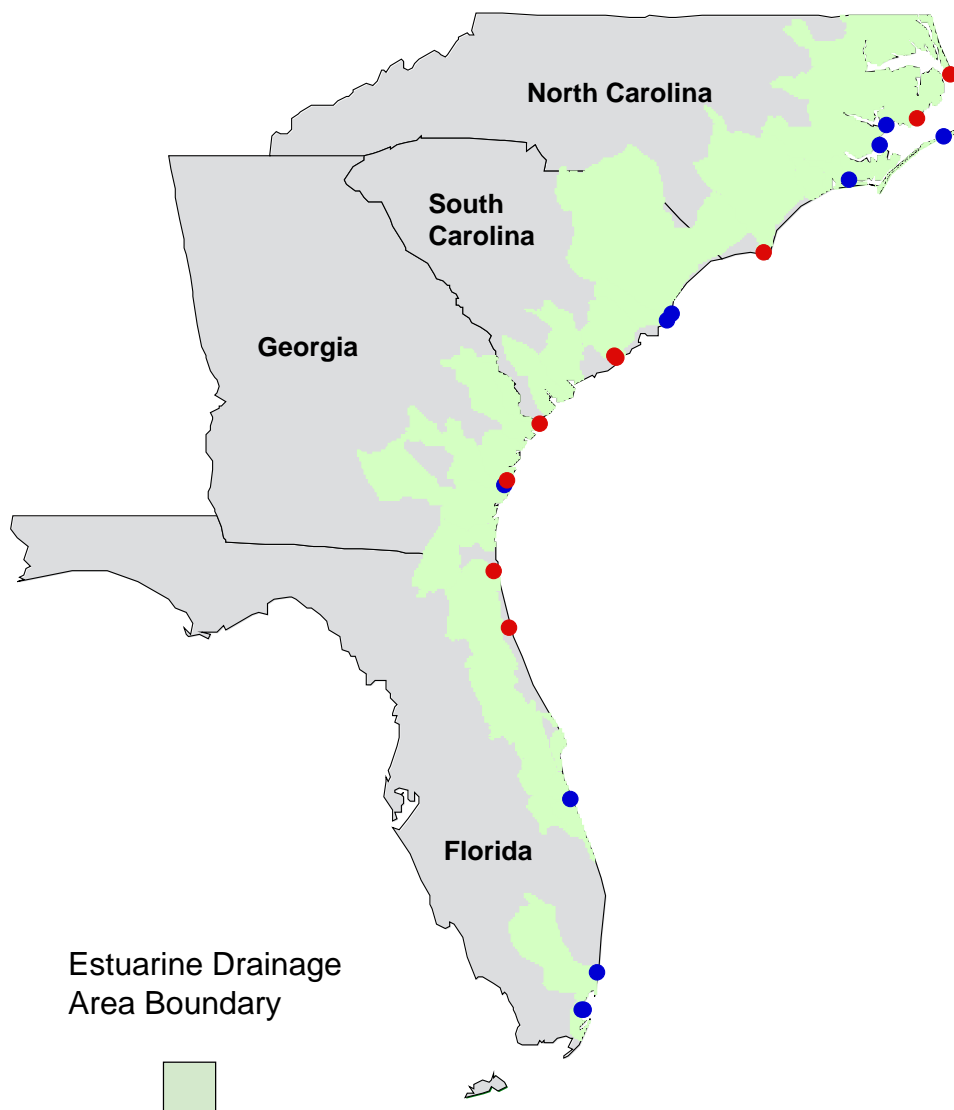
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South Atlantic Mussel Watch Sites

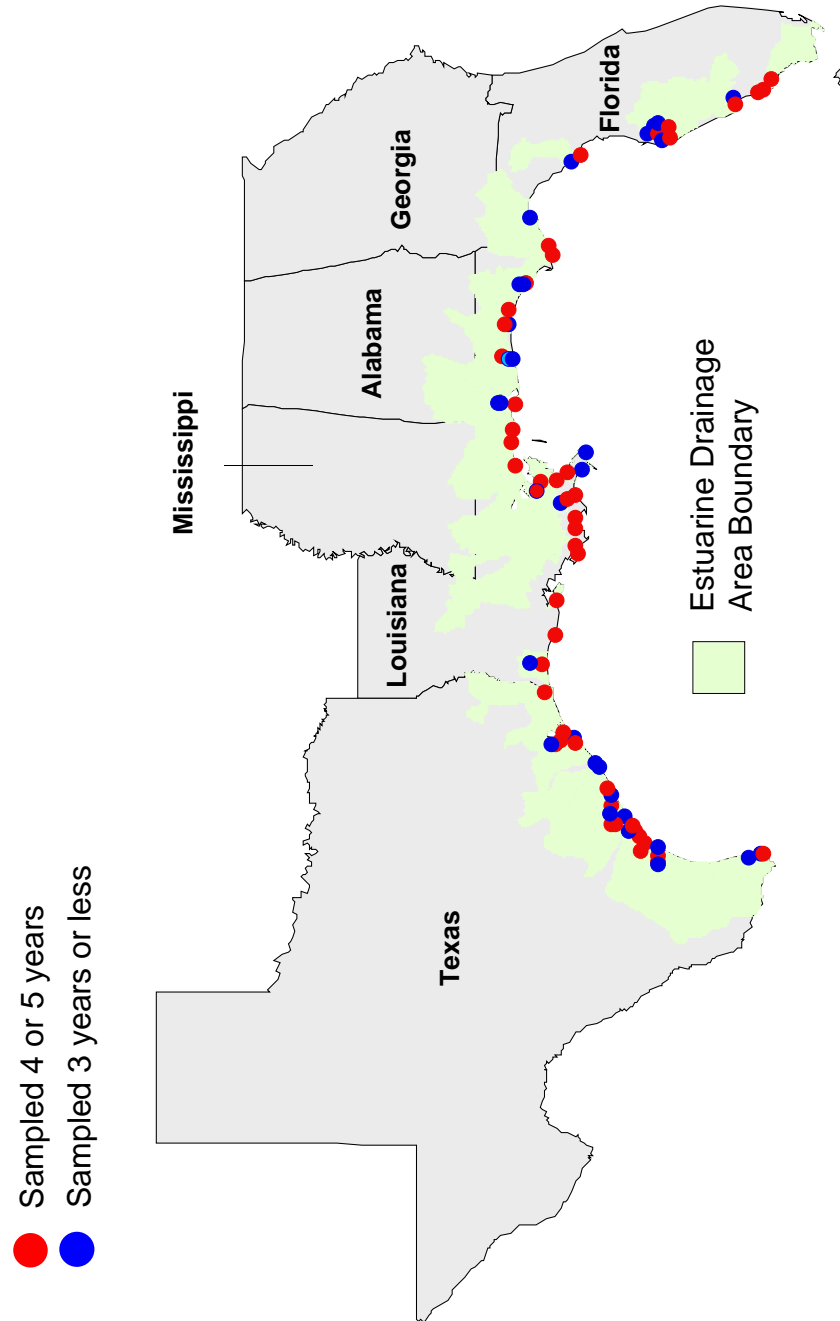
Figure 1c. *Mollusk collection sites 1986 through 1990.*

- Sampled 4 or 5 years
- Sampled 3 years or less



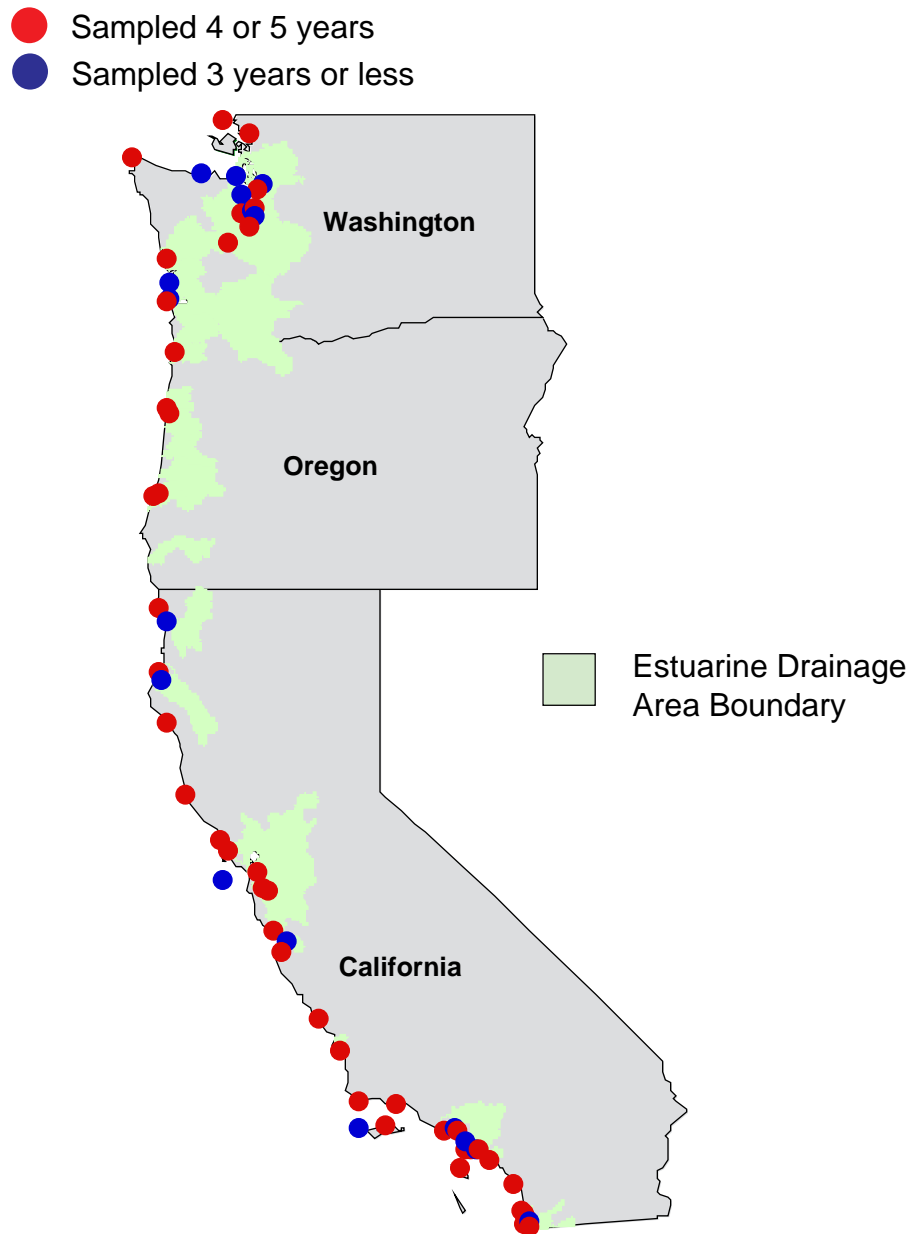
Gulf of Mexico Mussel Watch Sites

Figure 1d. *Mollusk collection sites 1986 through 1990*



Pacific Mussel Watch Sites

Figure 1e. *Mollusk collection sites 1986 through 1990.*



all sites are listed in Appendix A. The figures and Appendix A highlight 141 sites that were sampled in at least four of the five years between 1986 and 1990. Data from those sites are used to identify temporal trends in this report .

No single species of mollusk is common to all coasts. As a result, it has been necessary to collect four different ones: the mussel *Mytilus edulis* on the East Coast from Maine to Cape May, NJ; the oyster *Crassostrea virginica* from Delaware Bay southward and throughout the Gulf of Mexico; the mussels *M. edulis* and *M. californianus* on the West Coast; and the oyster *Ostrea sandvicensis* in Hawaii.

CHEMICALS MEASURED

The NS&T program monitors concentrations of trace metals and organic compounds. With the exception of chlorinated organic compounds such as DDT and PCB, which exist entirely as the result of human activities, a certain natural concentration of chemicals exists in mollusks even in the absence of human activity. Chemical concentrations exceeding natural levels should be considered “contamination,” and the exact line demarcating natural concentrations from contamination is not easily drawn. It depends on the species of mollusk itself as well as on many local and regional conditions.

Data on concentrations of the 10 trace metals and five groups of organic com-

pounds listed in Table 1 are used in this report to describe the distribution and trends of chemical concentrations in mollusks of the coastal and estuarine United States. Concentrations of each of these chemicals can serve as indicators of human activity. While the metals all have different uses, they can be categorized as chemicals that have been increasingly discharged to the environment as a result of industrialization.

The groups of organic compounds, however, cannot be categorized so generally. Two of the groups, total DDT (tDDT) and total chlordane (tCdane), are chlorinated pesticides. Use of DDT was banned in the United States in 1972. Chlordane use on U.S. crops ended in 1983, and its use for termite control effectively ended in 1988 (Shigenaka, 1990). Polychlorinated biphenyls (tPCB) are a mixture of chlorinated compounds first used in the 1920s for a number of industrial purposes. Their high heat capacities and low dielectric constants were exploited for use in electrical transformers and capacitors. PCB use in the United States began being phased out in 1971, and a ban on new uses took effect in 1976. Large changes in concentrations of tDDT and tPCB were seen at some locations in the 1970s following bans on further uses of tDDT and tPCB (Mearns et al., 1988), but the compounds are still found in tissues of organisms and marine sediments. PCB-containing devices are still in use, chlordane remains in the ground as a termiticide, and DDT remains in the environment because of its resistance to

degradation. The pesticide DDT is metabolized to DDE and DDD in the environment, but those compounds degrade very slowly under environmental conditions.

The three butyltin compounds, aggregated as tBT, are found in mollusks because tributyltin (TBT) has been used as an antifouling agent in the paint commonly used on ships and some underwater marine facilities. Its use on vessels under 75 feet long was banned in 1988. Tributyltin degrades to dibutyltin and then to monobutyl tin, which itself does not persist, so unlike the chlorinated compounds, tBT should degrade relatively quickly (Seligman et al., 1988). Consequently, the NS&T Program should find substantial decreases in tBT concentrations during the next several years.

Polycyclic aromatic hydrocarbons (PAHs) are similar to metals in the sense that they occur naturally. They are found in fossil fuels such as coal and oil. Their existence, however, is also attributable to humans because they are produced when organic matter is burned. A multitude of human activities, from coal and wood burning to waste incineration, create PAH compounds in excess of those that would exist naturally. In addition, human production, transport, and use of oil releases more PAHs to the environment, on a globally averaged basis, than does natural seepage. Because they are relatively more concentrated in oil than in combustion products, 2- and 3-ring

compounds, especially those with alkyl groups on a ring such as methyl- and dimethylnaphthalene and methylphenanthrene (Table 1), are sometimes classified separately from the higher molecular-weight 4- and 5-ring compounds. Since high concentrations of both types of compounds tend to be found in the same locations, all PAH compounds have been combined into a single group in this report.

All of these trace metals and groups of organic compounds can be acutely or chronically toxic to marine life and to humans under some conditions. On the other hand, while the elements arsenic, chromium, copper, nickel, selenium, and zinc can be toxic at high concentrations, they are also essential to the maintenance of life (Nielsen, 1988).

DIFFERENCES BETWEEN SPECIES IN CHEMICAL CONCENTRATIONS

One use of the Mussel Watch data is to compare chemical concentrations among sites. However, it is inappropriate to compare concentrations of some elements in mussels with those in oysters. This was demonstrated through analyses of mussels (*M. edulis*) and oysters (*C. virginica*) collected at one site in Long Island Sound. The results shown in Figure 2 indicate that, despite being exposed to the same environment, the species do not accumulate all chemicals to the same extent. Concentrations of copper, zinc, and silver are more than 10

Trace Metals

Arsenic (As)
Cadmium (Cd)
Chromium (Cr)
Copper (Cu)
Lead (Pb)

Nickel (Ni)
Mercury (Hg)
Selenium (Se)
Silver (Ag)
Zinc (Zn)

Organic Compounds*

Total DDT (tDDT)

The sum of concentrations of DDT (dichlorodiphenyltrichloroethane) and its metabolites DDE (dichlorodiphenyltrichloroethylene) and DDD (dichlorodiphenyldichloroethylene).

Total chlordane (tCdane)

The sum of concentrations of two major constituents of chlordane mixtures, cis-chlordane and trans-nonachlor and two minor components, heptachlor and heptachlorepoide.

Total polychlorinated biphenyls (tPCB)

The sum of the concentrations of di-, tri-, tetra-, penta-, hexa-, hepta-, octa-, and nonachlorobiphenyls. Since 1988, the equivalent tPCB has been calculated from the sum of concentrations of 18 individual PCB congeners.

Total polycyclic aromatic hydrocarbons (tPAH)

The sum of concentrations of 18 PAH compounds: six 2-ring compounds (biphenyl, naphthalene, 1-methylnaphthalene, 2-methylnaphthalene, 2,6-dimethylnaphthalene, and acenaphthene); four 3-ring compounds (fluorene, phenanthrene, 1-methylphenanthrene, and anthracene); three 4-ring compounds (flouranthene, pyrene, and benz[a]anthracene); and five 5-ring compounds (chrysene, benzo[a]pyrene, benzo[e]pyrene, perylene, and dibenz[a,h]anthracene).

Total butyl tin (tBT)

The sum of the concentrations of tributyl tin and its breakdown products dibutyl tin and monobutyl tin. (Concentrations in units of ng of Sn (as tBT)/g dry tissue.)

*The NS&T Program monitors concentrations of other chemicals, such as Dieldrin and Lindane, whose concentrations were below detection at more than 10% of the 1990 sites. Because such results make trend identification difficult, those compounds are not discussed in this report.

Table 1. *Chemicals measured in the National Status and Trends Program.*

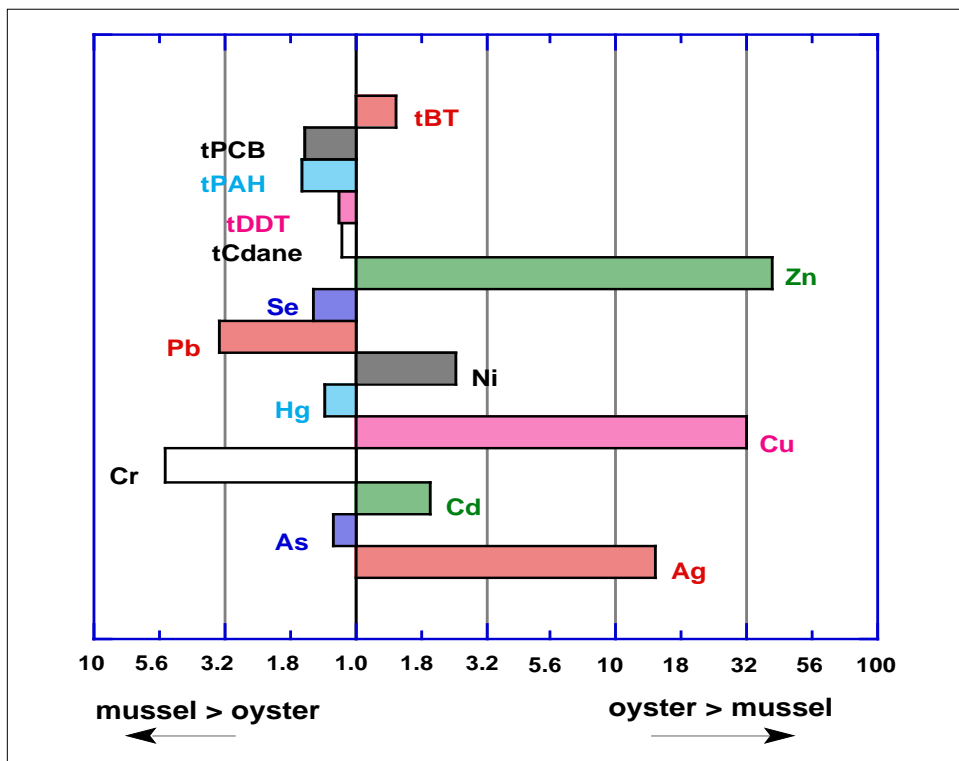


Figure 2. Factors by which concentrations in mussels differ from those in oysters at a single site.

times higher in oysters than in mussels, while the concentrations for chromium and lead are threefold higher in mussels. These large differences mean that concentrations of silver, copper, chromium, lead, and zinc in mussels cannot be compared with those in oysters. For the remaining chemicals, the differences are deemed small enough to ignore, and sites are compared regardless of species in this report. At a site near the mouth of the Columbia River, two species of mussels, *M. edulis* and *M. californianus*, have been sampled. Concentration differences were small for all chemicals,

and the two species can be considered equivalent.

In this regard, it is important to note that the primary reason for collecting and analyzing mollusks on a yearly basis is to track temporal trends in chemical concentrations. Annual data on concentrations of any chemical at a single site can be compared without consideration of the species, as long as the same species is collected every year. Consequently, while species differences do affect the analysis of national distributions of chemical concentrations, they

do not affect the analyses of trends based on changes at individual sites.

NATIONWIDE DISTRIBUTION OF CHEMICAL CONCENTRATIONS

In 1990, the Mussel Watch Project sampled 214 sites — the most sites sampled in any single year since the project began. Data from 1990 are used to describe the distribution of chemical concentrations throughout the Nation.

As the mercury and chlordane concentrations in Figure 3 illustrate, chemicals in mollusks at most sites are at the low end of the overall concentration range, and as concentrations increase they are found at fewer and fewer sites. This type of distribution was also found for chemical concentrations in sediments at NS&T sites (O'Connor, 1990), and is common among environmental data sets. As Figure 3 also exemplifies, when the logarithms of the concentrations are plotted, the distributions become bell-shaped and can be said to be “log-normal.” An advantage of this distribution is that it allows a statistically objective definition of a “high” concentration as one where the logarithmic value is more than the mean plus one standard deviation of the logarithms for all concentrations. As demonstrated in Figure 3, the mean and “high” concentrations for mercury and chlordane are 0.094 and 0.24 µg/g (dry) and 14 and 31 ng/g, respectively. For those and the other chemicals being evaluated in this report, the mean and “high” concentrations for

1990 are listed in Table 2.

This definition can be used to identify which Mussel Watch sites have mollusks with “high” concentrations of each chemical. Appendix A lists, in clockwise geographic sequence from Maine to Hawaii, all sites sampled from 1986 through 1990. It also indicates which chemicals, if any, had concentrations in the high range. Copper, zinc, silver, lead and chromium have been excluded because, as just discussed, concentration comparisons for those elements can only be done among sites with a common species.

On a national scale, high levels of organic contamination are clearly indicated in the urbanized areas of Boston, New York, Mobile, San Diego, San Francisco, and Los Angeles. There is also a tendency for concentrations to increase in these and other areas as sites are located closer to population centers. In Galveston Bay, for example, the higher concentrations are found near the ship channel to Houston rather than toward the Gulf of Mexico.

High concentrations associated with sites away from population centers are not all readily explained. Total butyltin (tBT) is high near marinas because it was used on boats, but high concentrations of other chemicals may be unrelated to human activity. High concentrations of cadmium in mussels collected along the coast of Northern California have been attributed to naturally high levels in

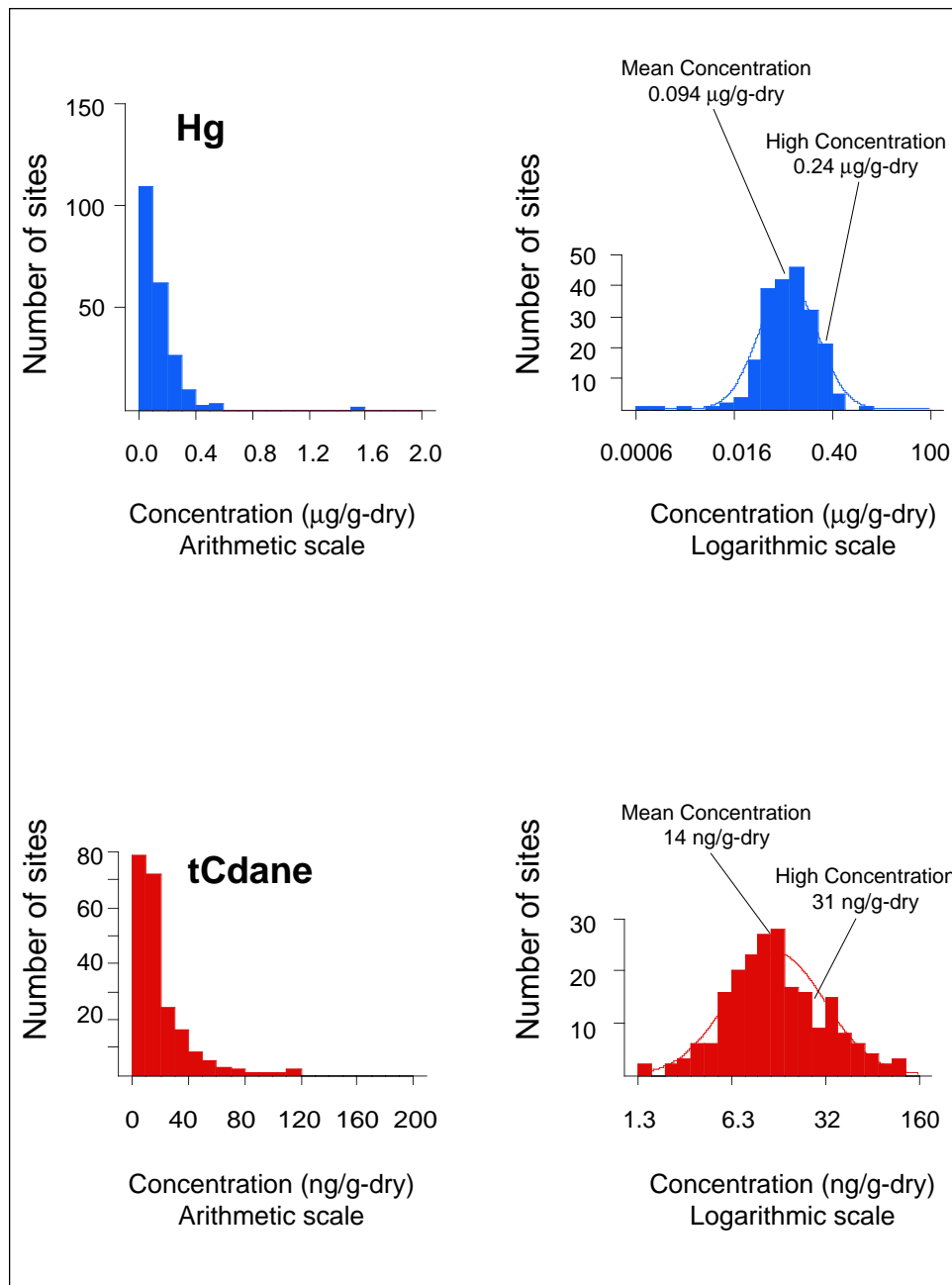


Figure 3. Distributions of mercury and total chlordane concentrations in mollusks on arithmetic and logarithmic scales.

Table 2. Geometric Mean and "High" concentrations^a from analyses of mollusks collected in 1990 at 214 sites (oysters collected at 107 sites and mussels at another 107).

Chemical	Geometric mean	"high"
arsenic	10 µg/g	17 µg/g
cadmium	2.7	5.7
mercury	0.094	0.24
nickel	1.7	3.3
selenium	2.5	3.5
tPCB	110 ng/g	470 ng/g
tDDT	37	120
tCdane	14	31
tPAH	260	890
tBT	81	350
(oysters only)		
silver	1.9 µg/g	3.7 µg/g
copper	150	360
zinc	2400	5200
lead	0.52	0.94
chromium	0.48	0.93
(mussels only)		
silver	0.17	0.58
copper	8.9	11
zinc	130	190
lead	1.8	4.3
chromium	1.7	3.0

^a All concentrations on a dry-weight basis of whole soft parts of mollusks. The "high" concentrations correspond to the mean plus one standard deviation of the logarithms of the individual site means.

deep-ocean water and upwelling of this water in that area (Goldberg et al., 1983). That supposition cannot be used to explain high cadmium concentrations in oysters at some Gulf of Mexico sites, however. Arsenic is distinctly high along the Southeast Coast — an observation that may be related to arsenic's natural association with phosphate, and the fact that phosphate deposits in that area are

sufficient to support mining activities.

The "high" levels have been statistically derived from the NS&T data for use in a relative sense to compare among sites. They cannot be related to biological effects. The U.S. Food and Drug Administration (FDA) has issued guidelines that warn against human consumption of shellfish with concentrations (on

a wet-weight basis) above specified levels of mercury and several chlorinated hydrocarbons. With one exception, none of the mollusks collected in the NS&T Program have had concentrations (adjusted to a wet-weight basis) that exceed those levels. The exception is the tPCB concentration in mussels collected at the Angelica Rock site in Buzzards Bay, MA (near New Bedford Harbor) in 1989, but not in prior years nor in 1990.

TEMPORAL TRENDS

Mussels and oysters are collected by the NS&T Program to monitor trends in chemical concentrations over time. Chemical concentrations in mussels and oysters are determined by the extent to which the organisms accumulate chemicals from the food they filter from their surrounding water and from the water itself. When chemical concentrations increase or decrease in their surroundings, the organisms are capable of increasing or decreasing the corresponding concentrations in their tissues (see for example, Roesijadi et al., 1984; Pruell et al., 1987). This, and the fact that they are immobile, make them ideal for monitoring changes in chemical concentrations at fixed sites.

Temporal trends can be defined through two approaches. First, a trend exists when a year-to-year change in the same direction occurs at the vast majority of sites. Secondly, there can be a statistically meaningful relationship between concentration and time. The butyltin

data provide an example of the first kind of trend. Detectable concentrations were found at 149 sites in both 1989 and 1990. Therefore, in 149 cases butyltin concentrations could have increased or decreased. While no importance can be attached to the direction of change at any *one* site, it is statistically significant that butyltin was lower at 103 sites in 1990. The probability of flipping a coin 149 times and getting 103 “heads” is very small. Similarly, there is practically no chance that 103 decreases in butyltin between 1989 and 1990 was a random event. Strictly speaking, the chance of that result occurring randomly is less than 1 in 1,000. Here, and throughout this report, results that are likely to occur randomly less than 5 times in 100 (0.05 level of significance) will be considered important.

Five years of data have been collected for the other chemicals. While the number of sites has increased over the years, comparisons between years will be limited to the 141 sites that were sampled in at least four of the five years. Applying the same test just applied to the tBT data (called the sign test) yields the results summarized in Table 3. There is no need to consider oysters and mussels separately for any chemical, since we are looking for differences at individual sites. Between 1989 and 1990, when butyltin was found to decrease, tPCB also decreased, and four chemicals — silver, arsenic, lead, and zinc — increased. Despite that,

Table 3. Results of sign test applied to mean chemical concentrations at the 141 sites sampled in at least four of the years between 1986 and 1990^a.

<u>Chem.</u>	<u>86-87</u>	<u>87-88</u>	<u>88-89</u>	<u>89-90</u>	<u>86-90</u>
Ag	-	-	-	Inc	-
As	Dec	-	Dec	Inc	-
Cd	Dec	-	-	-	Dec
Cr	Dec	-	-	-	-
Cu	-	-	-	-	-
Hg	-	-	-	-	-
Ni	-	Dec	Dec	-	Dec
Pb	Inc	-	Dec	Inc	-
Se	-	Inc	Dec	-	-
Zn	Dec	-	-	Inc	-
tPCB	Dec	-	-	Dec	Dec
tDDT	-	Dec	-	-	-
tCdane	Inc	Dec	-	-	Dec
tPAH	^b	Inc	Dec	-	^b
tBT				Dec	

^aComparisons made on a year-to-year basis and between 1986 and 1990. A statistically significant (0.05 level) proportion of changes in the increasing (Inc) or decreasing (Dec) direction are indicated, as are cases with no significant direction of change (-). Since it was not routinely measured in earlier years, tBt comparisons have been made for the 149 sites common to 1989 and 1990.

^b tPAH data for 1986 were not used. Geometric mean for that year was 430 ng/g which is 1.4 times the next highest annual geometric mean. Its high value may have been due to the analytical method used in that year for samples from the East and West Coasts. The last comparison for tPAH is an 87-90 comparison.

given the implementation of pollution controls and changes in industrial practices make any difference, decreases are expected. In fact, the overall conclusions from Table 3 are: (1) usually, between-year differences were not statistically in one direction; (2) when a direction was indicated, it was generally a decrease; and (3) when 1990 is compared with 1986, the only changes were decreases.

The sign test has been applied to the directions of concentration differences. Because no account has been taken of the amount of change, small and large differences have had equal bearing on results. To provide a sense of the magnitude of annual changes, the annual geometric means of chemical concentrations among all 141 sites sampled in four or five years are shown in Figures

4a to 4g. Here, since oyster sites and mussel sites are both part of the national mean, it has been necessary, again, to separate the data between species for silver, copper, zinc, chromium, and lead.

Decreases or increases identified in Table 3 also appear in Figure 4. The important conclusion from Figure 4 is that changes are small. For both elements and organic compounds, annual shifts in geometric means are generally less than 20 percent.

The sign test is useful when there are many comparisons to be made between any two years; it exploits the fact that there are many Mussel Watch sites. The

second method of trend detection identifies connections between concentration and time at individual sites and depends on the number of years of data. While plots will not be shown of annual mean concentrations of each chemical at each Mussel Watch site, this technique can be illustrated with two long-term data sets. Figure 5a shows the annual average carbon dioxide concentration in the atmosphere at the Mauna Loa Observatory in Hawaii for each year from 1959 through 1985 (Keeling and Boden, 1986). Figure 5b represents the annual mean temperature in the Northern Hemisphere from 1851 to 1984 relative to the average temperature over the years 1951 through 1970 (Jones et al., 1986).

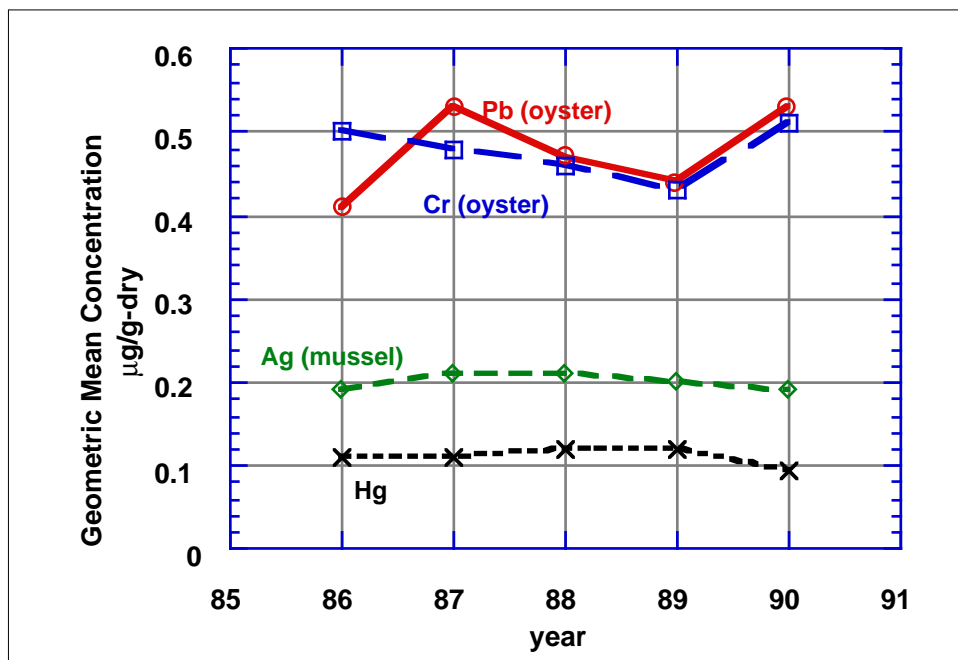


Figure 4a. Annual geometric mean concentrations of selected chemicals in mollusks.

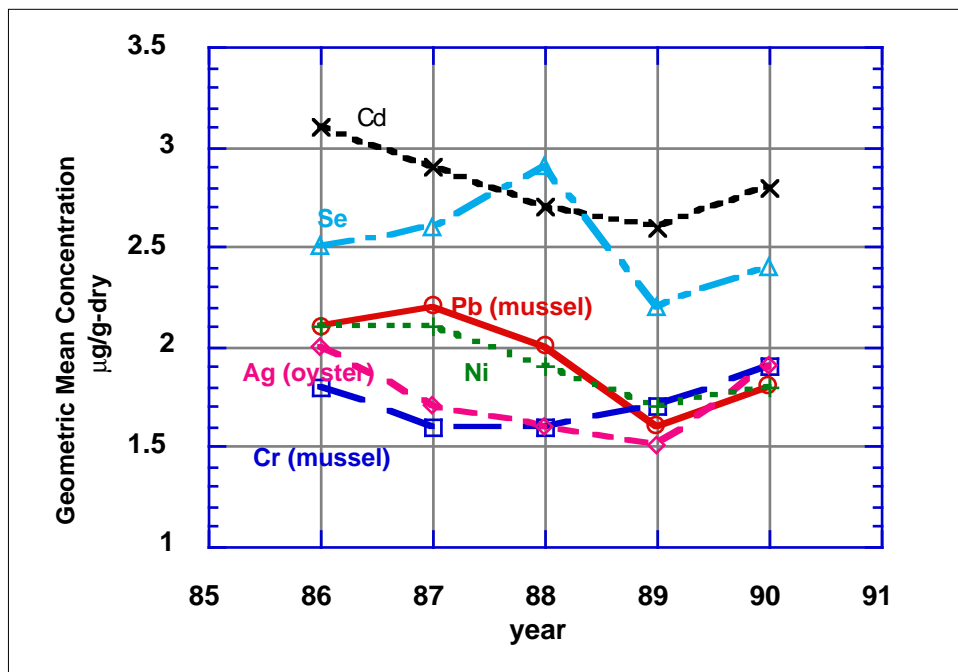


Figure 4b. Annual geometric mean concentrations of selected chemicals in mollusks.

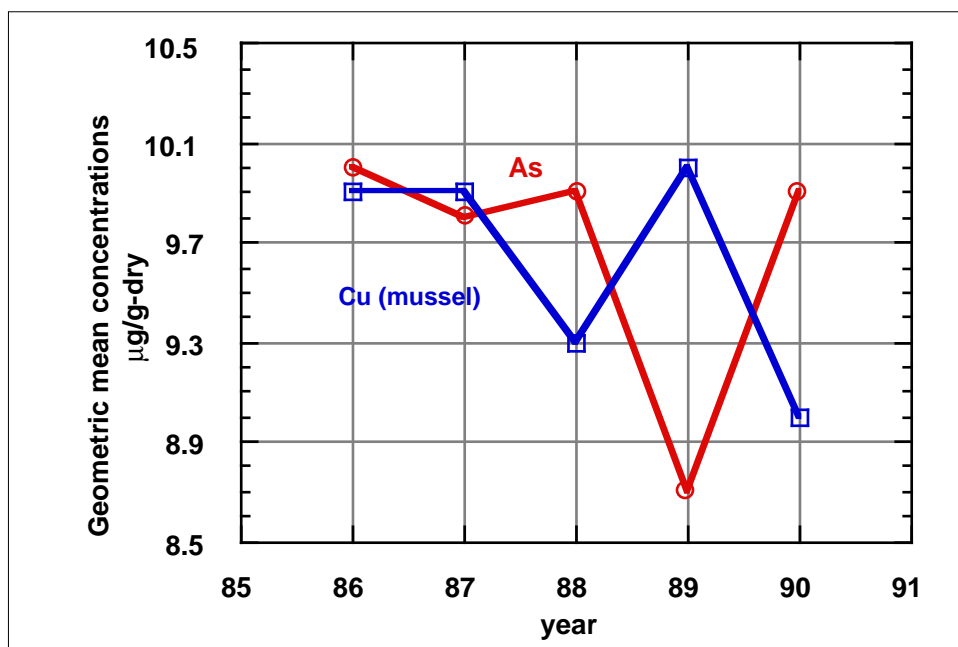


Figure 4c. Annual geometric mean concentrations of selected chemicals in mollusks.

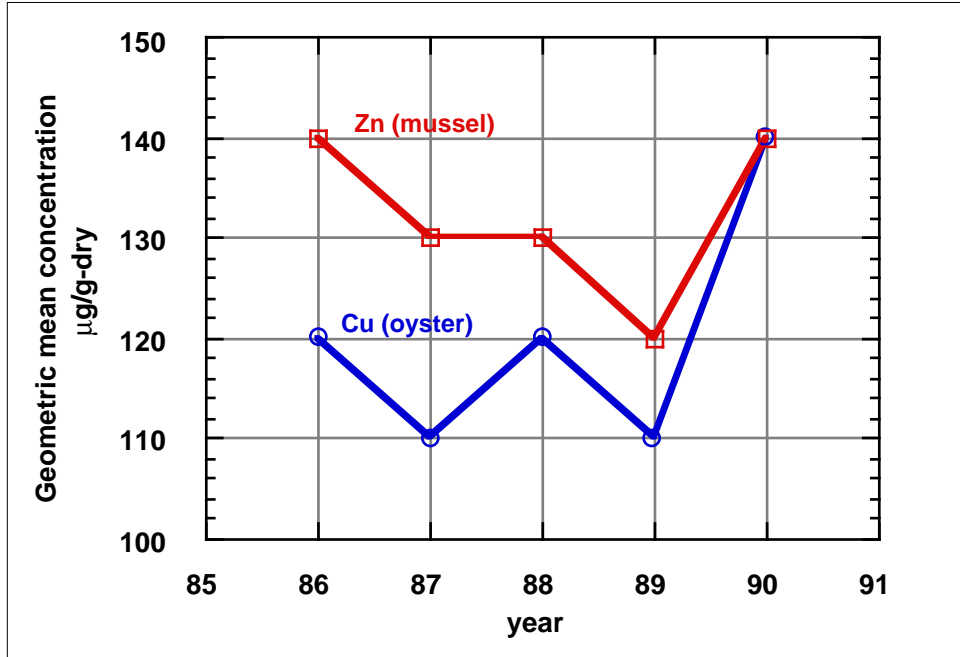


Figure 4d. Annual geometric mean concentrations of selected chemicals in mollusks.

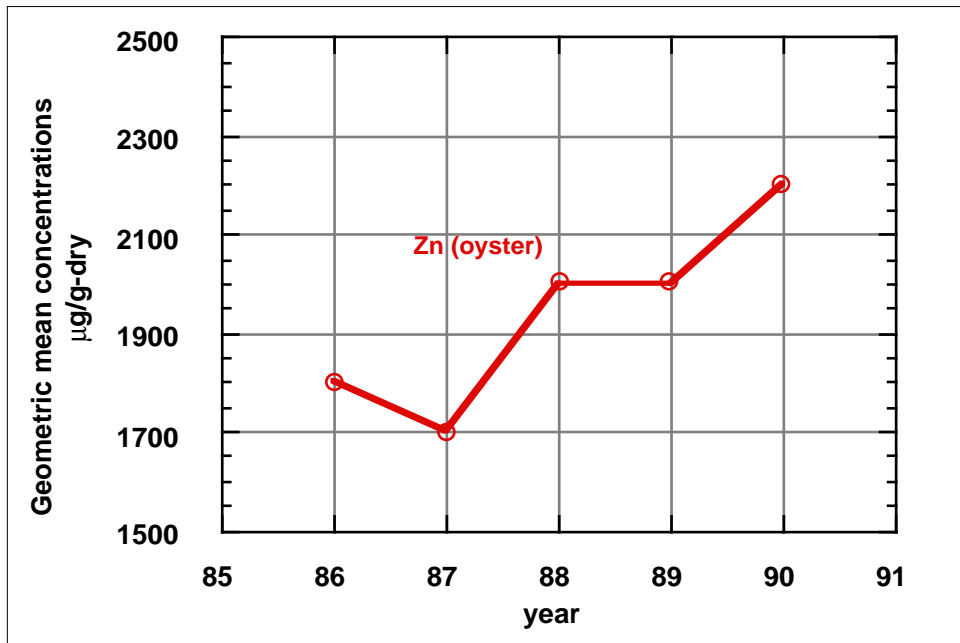


Figure 4e. Annual geometric mean concentrations of selected chemicals in mollusks.

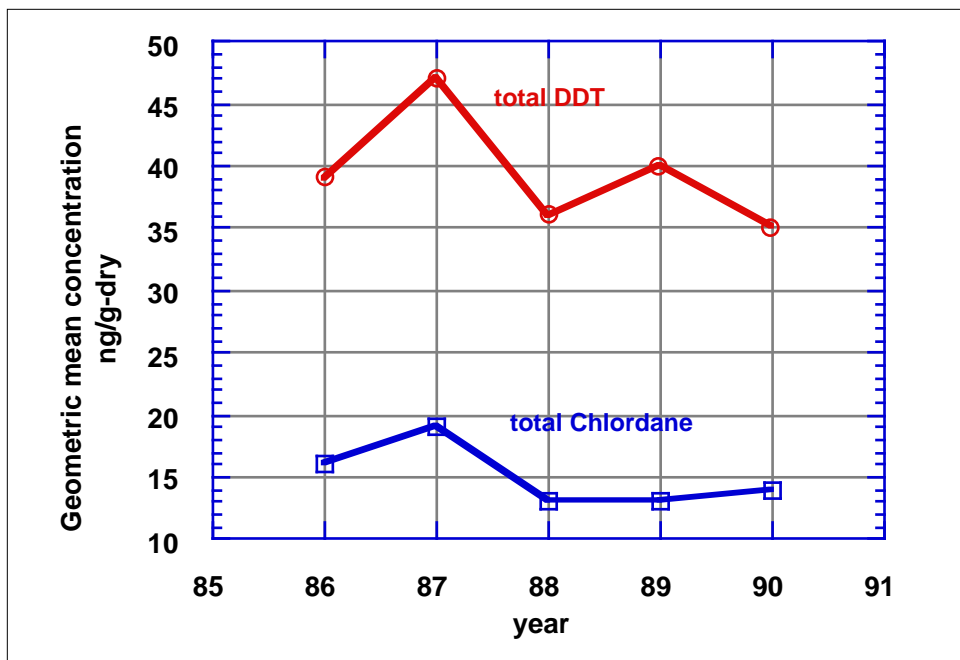


Figure 4f. Annual geometric mean concentrations of selected chemicals in mollusks.

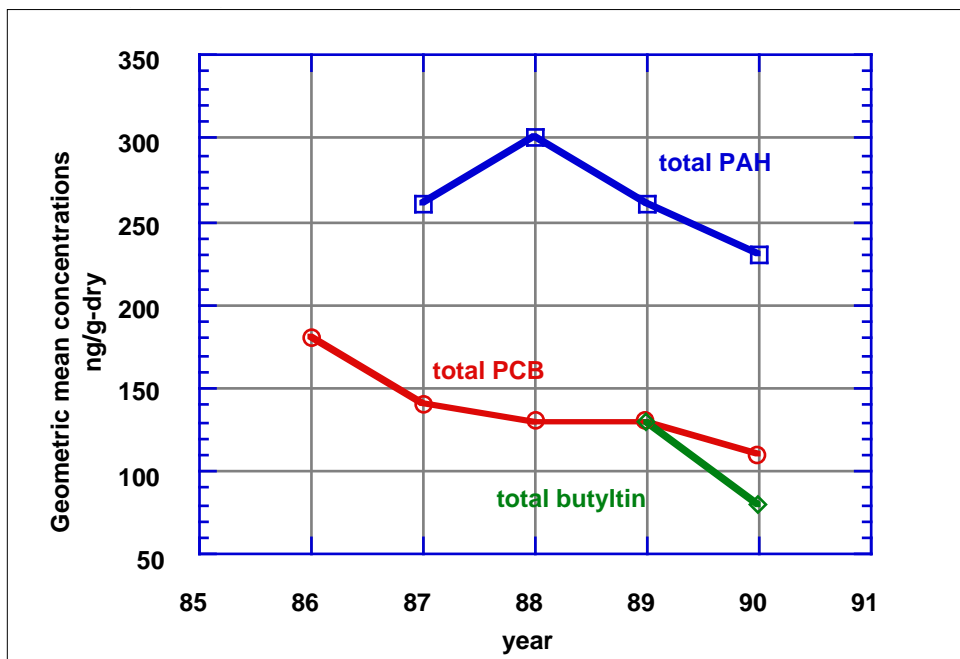


Figure 4g. Annual geometric mean concentrations of selected chemicals in mollusks.

A statistical attribute of the connection between concentration (or temperature) and time that constitutes a trend is the Spearman rank correlation coefficient. In the case of carbon dioxide in Figure 5a, the Spearman correlation coefficient is 1.0, as high a value as possible. The annual changes are small, but they occur invariably in the same direction. The temperature record is not so clear, because temperature in any one year is not always higher than the year before. Nevertheless, there is a trend in the temperature data. The Spearman correlation coefficient between temperature and year in Figure 5b is 0.57, and for a record with 134 observations the coefficient need equal only 0.14 for a statistically significant indication of a trend (at the 0.05 level). The temperature record

is “noisy” because many natural factors determine temperature in any given year, and extracting long-term trends requires data over many years. The carbon-dioxide record, on the other hand, is absolutely clear. The increase in carbon dioxide is attributable to the human activities of fossil-fuel burning and deforestation, and no natural factors are strong enough to complicate the record of those activities.

If all the Mussel Watch concentration time plots were shown, the vast majority would look like the temperature record in Figure 5b except they would only be for four or five years. With only five years of data, a trend cannot be identified unless the Spearman correlation is as high as 0.9. So to find a trend,

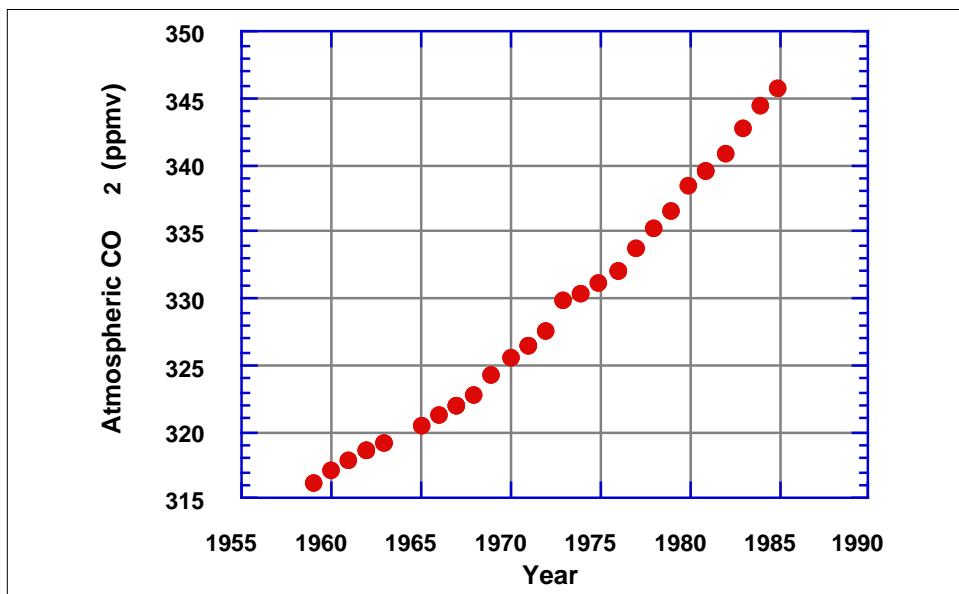


Figure 5a. Annual average atmospheric carbon dioxide concentration (Keeling and Boden, 1986).

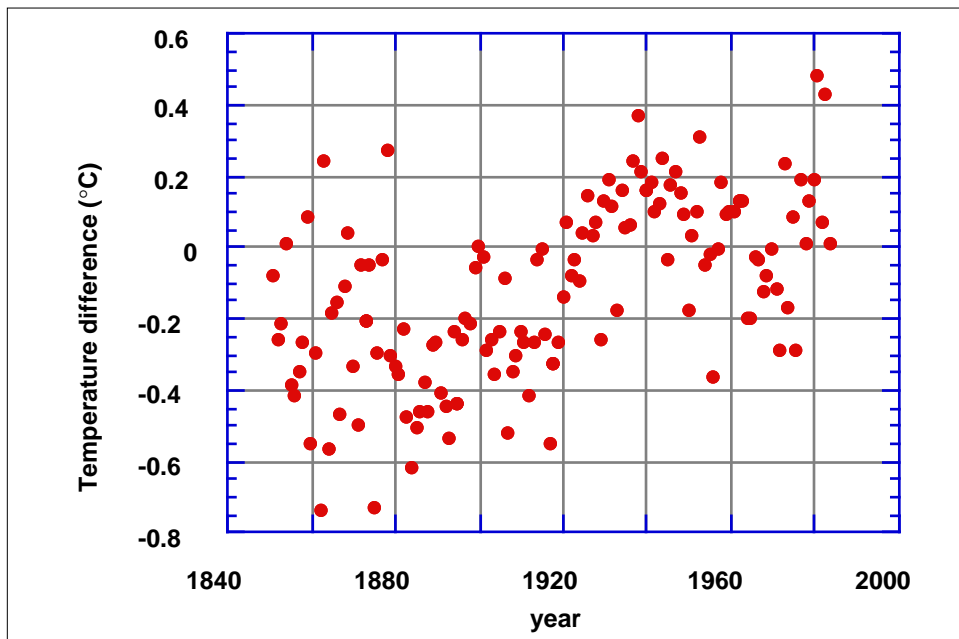


Figure 5b. *Difference between annual mean Northern Hemisphere air temperature and 20-year average of 1951 to 1970 (Jones et al., 1986).*

mean concentrations have to line up almost as perfectly as the carbon-dioxide concentrations in Figure 5a (i.e., the same direction of change occurs between each consecutive pair of years). Five years simply do not provide a long enough record to extract trends from noisy data.

The Spearman test was run on annual mean concentrations of 14 chemicals at the 141 sites with four or five years of data. Site-by-site and chemical-by-chemical results of this test are listed in Appendix B. Among 1,974 chemical-site combinations (14 chemicals x 141 sites), there are 239 cases (152 decreasing and 87 increasing) with strong cor-

relations between those concentrations and time. Finding trends in only 13% of the possible cases, reflects a lack of trends so strong that, like atmospheric carbon dioxide, they are evident with only a few years of data. There may be weaker trends that will be revealed with more years of data.

The statistical test allows for 5% of the correlations to be random occurrences rather than real connections between concentration and time. So 99 (0.05 x 1,974) of them may not be real. Nevertheless, of the 239 trends, the ones deserving close attention are those found among groups of sites. Among the nine Long Island Sound sites in Appendix B,

copper is decreasing at six, cadmium at five (Fig. 6a), silver at four, and zinc at three. Silver is decreasing at all four sites in Delaware Bay (Fig. 6b). Arsenic is decreasing (Fig. 6c) and zinc increasing at both sites in Terrebonne Bay. Just as finding clusters of sites with “high” concentrations (Appendix A) argues for those concentrations to be representative of an area, similar trends among sites in an area argues for the trend being real and area-wide.

NATURAL AND HUMAN INFLUENCES

If chemical concentrations in mollusks at any particular site increase because of human activities such as industry, agriculture, mining, or the wastes of daily living, the mollusks can be said to be chemically contaminated. If the chemical supply is purely natural, such as cadmium from deep ocean water bathing mollusks in Northern California, then concentrations do not represent contamination. For trace elements, there is no absolute way, based on concentration alone, to separate natural from human factors. An approximation of the extent of human influence at a site is the number of people in its proximity. Using 1990 census data (B. Davis, TIGER System Staff, U.S. Census Bureau), Spearman correlations were calculated between numbers of people residing within 20 km (12.4 miles) of each Mussel Watch site and chemical concentrations in mollusks collected in 1990. As expected, because their existence is en-

tirely or, to a large extent, due to human activity, the highest correlations were between population and concentrations of chlorinated organic compounds and PAHs. Statistically significant correlations were found between population and concentration for lead and zinc in both oysters and mussels, and for copper, mercury, and silver in mussels. There is no correlation between population and copper, mercury, or silver concentrations in oysters, and no or negative correlations between population and concentration of arsenic, cadmium, chromium, nickel, and selenium in either mussels or oysters.

A lack of correlation on a national scale, however, does not prove that concentrations are not affected by human activity at any individual site. There can well be human influences in rural areas. For example, the fact that mercury concentrations are high in oysters at two rural sites in Matagorda Bay, TX (Appendix A) is probably related to a reservoir of mercury contamination remaining from a major discharge of that element from a chlor-alkali plant in the 1970s (Holmes, 1977). Furthermore, there are rural sites with high concentrations of chlorinated organic compounds whose only source is human activity. Thus, it is not possible, simply on the basis of proximity to population centers, to separate human from natural effects on chemical concentrations in mollusks. This inability complicates trend detection since it is only the human-influenced component of chemical concentrations in mollusks

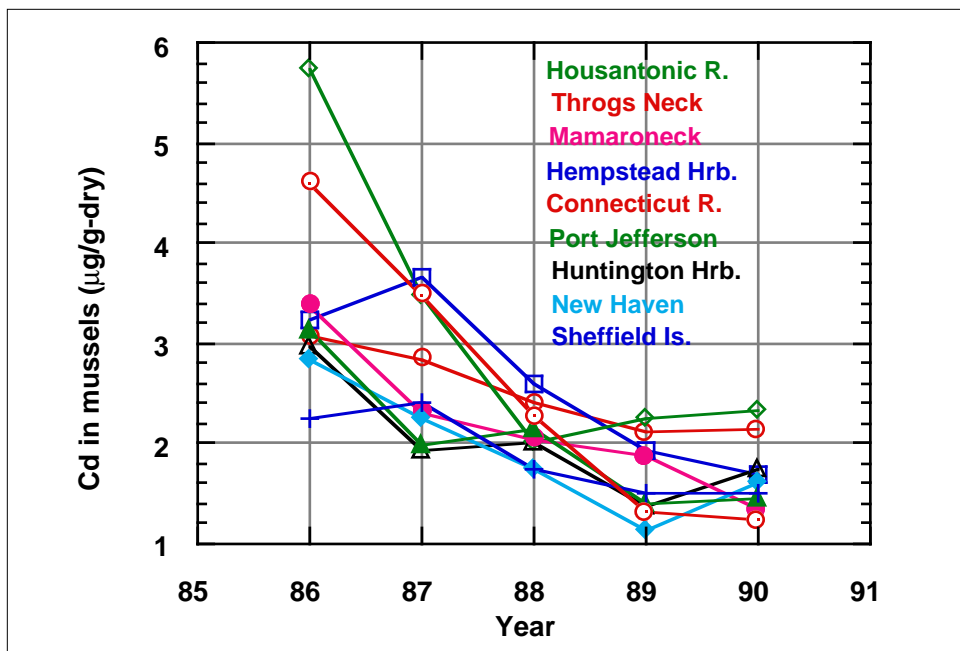


Figure 6a. Cadmium decreasing at Long Island Sound sites.

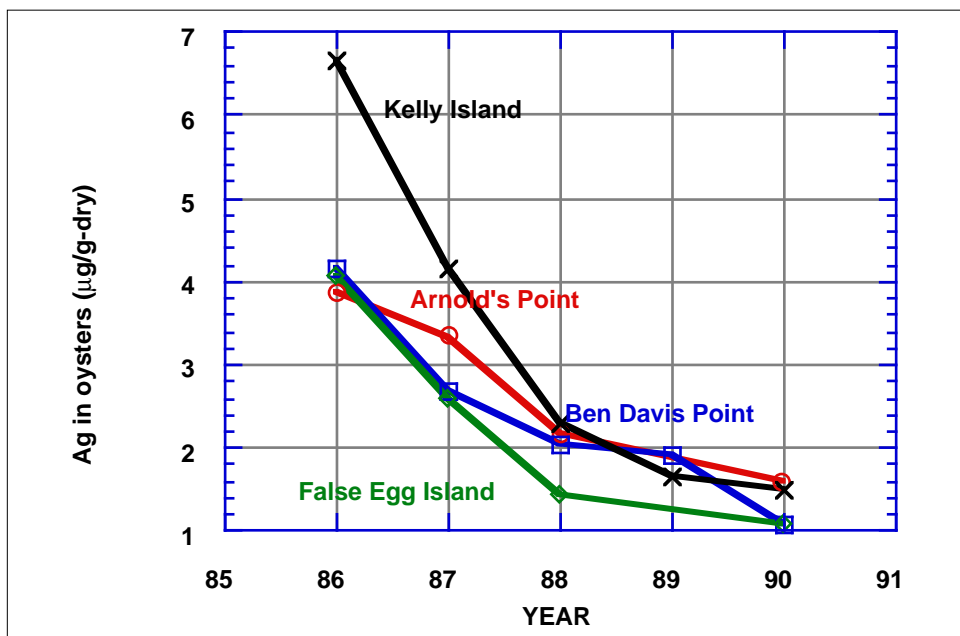


Figure 6b. Silver decreasing at Delaware Bay sites.

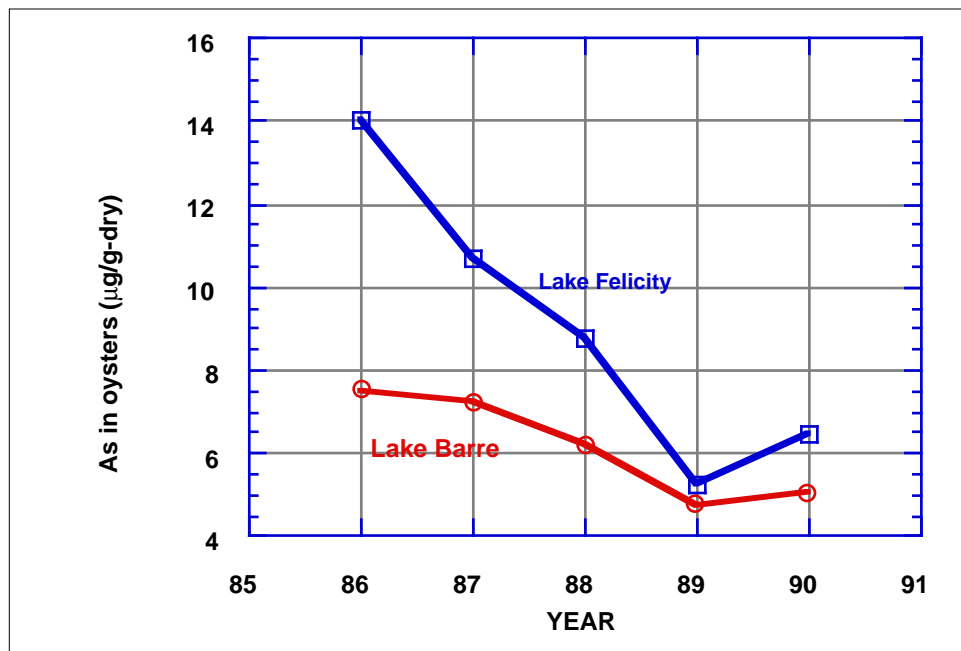


Figure 6c. Arsenic decreasing at Terrebonne Bay sites.

that can change in a consistent fashion with time.

There are natural factors, such as the difference between mussels and oysters, and salinity, and possibly reproductive state and season, that can affect concentrations (Phillips, 1980). Some of this is accounted for in the Mussel Watch Project by sampling in the same season every year, always collecting the same species at a site, and seeking mollusks of a certain size. Other factors, such as salinity, are monitored but cannot be controlled. There is ample reason for attributing interannual variations in concentration to natural factors, but it is difficult to attribute temporal

correlations with natural factors. The strong temporal correlations between concentrations and time that were found in the five-year data set do, most likely, imply a human influence at those sites.

LONGER-TERM TRENDS

Lauenstein et al. (1990) found decadal trends in lead concentrations by comparing NS&T data from 1986 through 1988 with data from analyses of mussels and oysters collected in 1976 through 1978 by a previous “mussel watch” program (Goldberg et al., 1983) sponsored by the U.S. Environmental Protection Agency (EPA). Fifty sites were common to both programs and, at 39 of them,

concentrations of lead were higher in the 1970s. The preponderance of change in that direction indicates a decreasing trend that was attributed to the phaseout of leaded gasoline. Yet, when the Sign test was applied to Mussel Watch data between 1986 and 1990 at 141 sites (Table 3), it failed to detect a trend. This may be due to the fact that the major effect of human intervention occurred in the 1970s, and that trends occurring in the 1980s are smaller and more easily masked by natural effects on interannual variation.

Large annual changes in past decades, followed by smaller shifts in the present day, are common in concentrations of organochlorine compounds. Figure 7, from O'Connor (1990), is a 19-year record of tPCB in mussels at the Mussel Watch site off Royal Palms Park on the Palos Verdes coast of Los Angeles. It is based on three sets of data, including that from the NS&T Program. It shows a dramatic decrease that began in 1971 when PCB use began to be phased out in the United States. The magnitude of the decrease may be magnified by the site's location, within 10 km of a major sewage outfall, but other cases have also documented temporal decreases in chlorinated organic contamination. Sericano et al. (1990) have combined data from diverse sources to show historical decreases in the average tDDT concentration in oysters in the Gulf of Mexico. Suns et al. (1991) found decreases in concentrations of tDDT and tPCB in fish collected in Lake Ontario when data from the mid-

1970s were compared with data from the 1980s. Robinson et al. (1990) examined data from analyses of human fat tissues collected from autopsies or from surgical patients in 1972 through 1983, and found a large decrease in the early 70s, followed by a more-or-less steady decrease to 1983, in the percentage of tissue samples containing more than 1,000 ng/g of tPCB. All of these reports reveal long-term decreases in tPCB in the tissues of organisms, but in all cases, the decreases are made evident over decadal time scales.

CONCLUSIONS

Data from the NOAA NS&T Mussel Watch Project show more decreases than increases in chemical concentrations between 1986 and 1990. At most individual sites there are no strong correlations between concentration and year, but where correlations are found decreases outnumber increases. This tendency for contamination to decrease is occurring at the same time that our society is taking more and more steps to control pollution. It supports the view that the imposition of control technologies and other recent actions to limit contaminant releases have, in general, stemmed the increasing levels of chemical contaminants previously observed in our coastal and estuarine waters. However, with only five years of data, it has not been possible to clearly establish long-term trends and more years of sampling are required to distinguish more

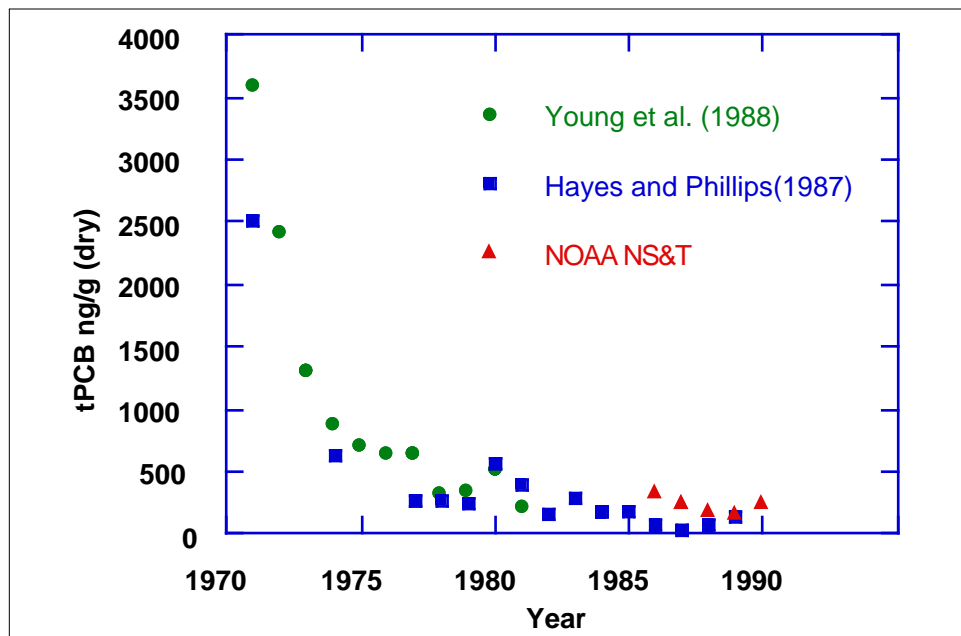


Figure 7. Annual concentration of total PCB in mussels at NS&T site at Palos Verdes, CA.

clearly the effects of human activity from natural influences on chemical concentrations.

ACKNOWLEDGMENTS

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Appendix A

This appendix lists all 234 sites sampled in the NS&T Mussel Watch Project. The most recent year of sampling (up to 1990) is listed in the column headed "yr." Concentrations in that year have been compared with "high" concentrations listed in Table 2, and a dot (•) in columns headed by symbols for chemicals indicates that the mean concentration exceeded the "high" value. Sites that have been sampled in at least four years between 1986 and 1990 are printed in bold type. The species sampled (column Sp) are the mussel *Mytilus edulis* ("me"), the oyster *Crassostrea virginica* ("cv"), the mussel *M. californianus* ("mc"), and the oyster *Ostrea sandvicensis* ("os"). The numbers of people residing within 20 km of each site according to the 1990 census are categorized as 1 through 6 under the heading "pop" by this scheme: category 1 for less than 10,000; 2 for 10,000 to 50,000; 3 for 50,000 to 100,000; 4 for 100,000 to 500,000; 5 for 500,000 to 1,000,000; and 6 for more than 1,000,000.

Appendix A. Roster of Mussel Sites and Chemicals at "High" Concentrations.

Yr	Sp	Pop	Main Location	Specific Location	St	As	Cd	Hg	Ni	Se	tPCB	tDDT	tCdne	tPAH	tBT
			NORTH ATLANTIC												
90	me	2	Penobscot Bay	Sears Island	ME			•							
90	me	1	Penobscot Bay	Pickering Island	ME										
90	me	2	Merriconeag Sound	Stover Point	ME										
90	me	3	Cape Arundel	Kennebunkport	ME										
90	me	2	Cape Ann	Gap Head	MA	•		•							•
90	me	4	Salem Harbor	Folger Point	MA										
90	me	5	Massachusetts Bay	Nahant Bay	MA			•							
90	me	6	Boston Harbor	Deer Island	MA			•		•	•	•	•	•	
90	me	6	Boston Harbor	Dorchester Bay	MA					•	•	•	•	•	
90	me	5	Boston Harbor	Hingham Bay	MA			•		•					
90	me	6	Boston Harbor	Brewster Island	MA			•							
90	me	4	Duxbury Bay	Clarks Island	MA										
90	me	4	Massachusetts Bay	North River	MA			•							
			MID-ATLANTIC												
90	me	2	Cape Cod	Nauset Harbor	MA										
90	me	4	Buzzards Bay	Cape Cod Canal	MA	•									
90	me	3	Buzzards Bay	Naushon Island	MA	•					•				
90	me	3	Buzzards Bay	West Falmouth	MA	•					•				
90	me	4	Buzzards Bay	Round Hill	MA	•					•				
90	me	4	Buzzards Bay	Angelica Rock	MA						•				

Yr	Sp	Pop	Main Location	Specific Location	St	As	Cd	Hg	Ni	Se	tPCB	tDDT	tCdne	tPAH	tBT
90	me	3	Buzzards Bay	Goosebury Neck	MA	•				•					
90	me	5	Narragansett Bay	Patience Island	RI								•		
90	me	4	Narragansett Bay	Dyer Island	RI				•						
89	me	4	Narragansett Bay	Dutch Island	RI				•						
90	me	1	Block Island	Block Island	RI										
90	me	2	Long Island Sound	Gardiners Bay	NY										
90	me	3	Long Island Sound	Connecticut River	CT					•					
90	me	5	Long Island Sound	New Haven	CT					•			•		
90	me	4	Long Island Sound	Housatonic River	CT					•			•		
90	me	4	Long Island Sound	Sheffield Island	CT								•		
90	me	5	Long Island Sound	Huntington Harbor	NY								•		
90	me	4	Long Island Sound	Port Jefferson	NY										•
90	me	6	Long Island Sound	Mamaroneck	NY					•			•		
90	me	6	Long Island Sound	Hempstead Harbor	NY								•		
90	me	6	Long Island Sound	Throgs Neck	NY					•			•		
90	me	4	Moriches Bay	Tuthill Point	NY			•							
90	me	5	Long Island	Fire Island Inlet	NY			•		•					
90	me	6	Long Island	Jones Inlet	NY			•							
90	me	6	Hud./Rar. Estuary	Jamaica Bay	NY					•			•		
90	me	6	Hud./Rar. Estuary	Upper Bay	NY			•		•			•		•
90	me	6	Hud./Rar. Estuary	Lower Bay	NY			•		•			•		•
90	me	6	Hud./Rar. Estuary	Raritan Bay	NJ					•			•		
90	me	6	New York Bight	Sandy Hook	NJ			•		•			•		•
90	me	4	New York Bight	Long Branch	NJ			•		•			•		
90	me	4	New York Bight	Shark River	NJ			•		•			•		•

Yr	Sp	Pop	Main Location	Specific Location	St	As	Cd	Hg	Ni	Se	tPCB	tDDT	tCdne	tPAH	tBT
90	me	3	Barnegat Inlet	Barnegat Light	NJ										
90	me	4	Absecon Inlet	Atlantic City	NJ										
90	me	3	Delaware Bay	Cape May	NJ					•					
90	cv	2	Delaware Bay	False Egg Island Pt.	NJ				•		•				
90	cv	2	Delaware Bay	Ben Davis Pt. Shoal	NJ				•		•				•
90	cv	2	Delaware Bay	Arnolds Point Shoal	NJ		•		•		•				
89	cv	2	Delaware Bay	Hope Creek	NJ		•		•		•				
89	cv	2	Delaware Bay	Woodland Beach	DE		•		•		•				
90	cv	3	Delaware Bay	Kelly Island	DE				•		•				
90	me	2	Delaware Bay	Cape Henlopen	MD					•					
90	cv	4	Chesapeake Bay	Bodkin Point	MD		•		•				•		•
90	cv	4	Chesapeake Bay	Mountain Point Bar	MD		•		•						•
90	cv	4	Chesapeake Bay	Hackett Point Bar	MD				•		•				
90	cv	2	Chesapeake Bay	Choptank River	MD				•						
90	cv	3	Chesapeake Bay	Hog Point	MD				•						
90	cv	2	Potomac River	Ragged Point	VA				•						
90	cv	2	Potomac River	Mattox Creek	VA				•						
90	cv	2	Potomac River	Swan Point	MD				•						
87	cv	2	Chesapeake Bay	Ingram Bay	VA										
90	cv	2	Rappahannock River	Ross Rock	VA		•								
90	cv	1	Chesapeake Bay	Cape Charles	VA										
90	cv	4	Chesapeake Bay	Dandy Point	VA										•
90	cv	4	Chesapeake Bay	James River	VA		•		•						•
90	cv	2	Chincoteague Bay	Chincoteague Inlet	VA										

Y r	Sp	Pop	Main Location	Specific Location	St	As	Cd	Hg	Ni	Se	tPCB	tDDT	tCdne	tPAH	tBT
			SOUTH ATLANTIC												
90	cv	2	Quinby Inlet	Upshur Bay	VA				•						
90	cv	1	Pamlico Sound	Pungo River	NC										
90	cv	2	Roanoke Sound	John Creek	NC										
90	cv	1	Pamlico Sound	Wysocking Bay	NC										
90	cv	1	Pamlico Sound	Neuse River	NC										
90	cv	1	Pamlico Sound	Cape Hatteras	NC										
90	cv	2	Cape Fear	Battery Island	NC	•									
90	cv	2	Beaufort Inlet	Pivers Island	NC	•								•	
90	cv	2	Winyah Bay	Lower Bay	SC	•			•						
90	cv	1	Santee River	North Bay	SC	•			•						
90	cv	4	Charleston Harbor	Fort Johnson	SC	•									
90	cv	4	Charleston Harbor	Shutes Folly Island	SC	•			•						
90	cv	3	Savannah R. Estuary	Tybee Island	GA	•									
90	cv	1	Sapelo Sound	Sapelo Island	GA	•									
90	cv	2	Altamaha River	Wolfe Island	GA	•			•						
90	cv	4	St. Johns River	Chicopit Bay	FL	•			•						•
90	cv	2	Matanzas River	Crescent Beach	FL										
90	cv	2	Indian River	Sebastian River	FL									•	•
90	cv	6	North Miami	Maule Lake	FL										•
90	cv	4	Biscayne Bay	Gould's Canal	FL	•									•
87	cv	1	Biscayne Bay	Princeton Canal	FL									•	

Y r	Sp	Pop	Main Location	Specific Location	St	As	Cd	Hg	Ni	Se	tPCB	tDDT	tCdre	tPAH	tBT
			GULF OF MEXICO												
90	cv	1	Everglades	Faka Union Bay	FL										
90	cv	3	Rookery Bay	Henderson Creek	FL										
90	cv	4	Naples Bay	Naples Bay	FL										•
90	cv	4	Charlotte Harbor	Bird Island	FL			•							
90	cv	4	Charlotte Harbor	Fort Meyers	FL								•		
90	cv	4	Tampa Bay	Mullet Key Bayou	FL										
90	cv	4	Tampa Bay	Cockroach Bay	FL								•		
90	cv	5	Tampa Bay	Navarez Park	FL			•					•		
88	cv	4	Tampa Bay	Hillsborough Bay	FL			•							
90	cv	5	Tampa Bay	Papys Bayou	FL			•							
90	cv	5	Tampa Bay	Peter O. Knight Airport	FL								•		•
90	cv	5	Tampa Bay	Old Tampa Bay	FL				•						
90	cv	1	Cedar Key	Black Point	FL			•							
88	cv	1	Suwannee River	West Pass	FL										
90	cv	2	Apalachee Bay	Spring Creek	FL			•							
90	cv	1	Apalachicola Bay	Cat Point Bar	FL										
90	cv	1	Apalachicola Bay	Dry Bar	FL										
90	cv	4	Panama City	Little Oyster Bar	FL							•			
90	cv	4	Panama City	Municipal Pier	FL			•						•	•
90	cv	4	St. Andrew Bay	Watson Bayou	FL									•	
90	cv	1	Choctawhatchee Bay	Off Santa Rosa	FL			•		•					
90	cv	4	Choctawhatchee Bay	Postil Point	FL			•				•	•		
90	cv	4	Choctawhatchee Bay	Joe's Bayou	FL			•						•	
90	cv	4	Pensacola Bay	Sabine Point	FL										

Y r	Sp	Main Location	Specific Location	St	As	Cd	Hg	Ni	Se	tPCB	tDDT	tCdre	tPAH	tBT
90	cv	4 Pensacola Bay	Public Harbor	FL										
89	cv	4 Pensacola Bay	Indian Bayou	FL										
90	cv	4 Mobile Bay	Dog River	AL		•				•	•	•	•	
90	cv	4 Mobile Bay	Hollingers Island Chan.	AL		•				•	•	•	•	
90	cv	2 Mobile Bay	Cedar Point Reef	AL						•				
90	cv	3 Mississippi Sound	Pascagoula Bay	MS	•									
90	cv	4 Mississippi Sound	Biloxi Bay	MS								•	•	
90	cv	3 Mississippi Sound	Pass Christian	MS		•								
88	cv	4 Lake Borgne	New Orleans	LA				•	•					
90	cv	1 Lake Borgne	Malheureux Point	LA		•	•							
90	cv	1 Breton Sound	Bay Gardene	LA										
90	cv	1 Breton Sound	Sable Island	LA		•								
90	cv	1 Mississippi River	Tiger Pass	LA		•								
90	cv	1 Mississippi River	Pass a Loutre	LA		•								
90	cv	1 Barataria Bay	Bayou Saint Denis	LA										
88	cv	1 Barataria Bay	Turtle Bay	LA										
90	cv	1 Barataria Bay	Middle Bank	LA									•	•
90	cv	1 Terrebonne Bay	Lake Felicity	LA										
90	cv	1 Terrebonne Bay	Lake Barre	LA										
90	cv	3 Caillou Lake	Caillou Lake	LA										
90	cv	1 Atchafalaya Bay	Oyster Bayou	LA		•								
90	cv	1 Vermillion Bay	Southwest Pass	LA		•						•		
90	cv	1 Joes Harbor Bayou	Joseph Harbor Bay	LA					•					
90	cv	1 Calcasieu Lake	Lake Charles	LA										
90	cv	1 Calcasieu Lake	St. Johns Island	LA		•			•					

Y r	Sp	Main Location	Specific Location	St	As	Cd	Hg	Ni	Se	tPCB	tDDT	tCdne	tPAH	tBT
90	cv	3 Sabine Lake	Blue Buck Point	TX					•					
90	cv	2 Galveston Bay	Hanna Reef	TX					•					
90	cv	4 Galveston Bay	Ship Channel	TX					•	•			•	•
90	cv	4 Galveston Bay	Yacht Club	TX								•		•
90	cv	4 Galveston Bay	Todd's Dump	TX										
90	cv	4 Galveston Bay	Confederate Reef	TX										
90	cv	4 Galveston Bay	Offatts Bayou	TX								•		
90	cv	3 Brazos River	Freeport Surfside	TX					•		•			
90	cv	2 Brazos River	Cedar Lakes	TX										
90	cv	1 Matagorda Bay	East Matagorda	TX		•			•					
88	cv	1 Matagorda Bay	Dog Island	TX					•					
90	cv	1 Matagorda Bay	Carancahua Bay	TX		•								
90	cv	1 Matagorda Bay	Tres Palacios Bay	TX		•			•					
89	cv	2 Matagorda Bay	Gallinipper Point	TX			•							
90	cv	2 Matagorda Bay	Lavaca River Mouth	TX		•	•							
90	cv	1 Espiritu Santo	South Pass Reef	TX		•								
90	cv	1 Espiritu Santo	Bill Days Reef	TX										
87	cv	1 San Antonio Bay	Mosquito Point	TX					•					
90	cv	1 San Antonio Bay	Panther Point Reef	TX		•			•					
90	cv	1 Mesquite Bay	Ayres Reef	TX		•			•					
90	cv	2 Copano Bay	Copano Reef	TX		•			•					
88	cv	2 Aransas Bay	Harbor Island	TX					•				•	
90	cv	2 Aransas Bay	Long Reef	TX		•								
90	cv	4 Corpus Christi	Boat Harbor	TX		•			•					•
90	cv	4 Corpus Christi	Ingleside Cove	TX										

Yr	Sp	Main Location	Specific Location	St	As	Cd	Hg	Ni	Se	tPCB	tDDT	tCdne	tPAH	tBT
90	mc	1 Point Conception	Point Conception	CA										
90	mc	4 San Luis Ob. Bay	Point San Luis	CA										
90	mc	4 San Simeon Point	San Simeon Point	CA	•	•		•						
90	mc	4 Pacific Grove	Lovers Point	CA	•	•								
90	mc	4 Monterey Bay	Moss Landing	CA						•		•		
90	mc	4 Monterey Bay	Point Santa Cruz	CA										
88	mc	1 Farallon Islands	East Landing	CA										
90	me	5 San Francisco Bay	Dumbarton Bridge	CA	•	•	•	•	•	•	•	•		
90	me	5 San Francisco Bay	San Mateo Bridge	CA	•	•		•	•	•	•	•		
90	me	6 San Francisco Bay	Emeryville	CA	•	•	•	•	•	•	•	•	•	•
90	me	1 Tomales Bay	Spenger's Residence	CA				•						
90	mc	2 Bodega Bay	Bodega Bay Entrance	CA	•	•								
90	mc	2 Point Arena	Lighthouse	CA	•	•		•						
90	mc	1 Point Delgada	Shelter Cove	CA	•	•		•						
90	mc	3 Eureka	Samoa Bridge	CA				•						
90	mc	3 Humboldt Bay	Jetty	CA										
89	mc	1 Klamath River	Flint Rock Head	CA				•						
90	mc	2 Point St. George	Point St. George	CA	•	•		•						
90	mc	2 Coos Bay	Coos Head	OR										•
90	me	2 Coos Bay	Russell Point	OR										
90	me	2 Yaquina Bay	Oneatta Point	OR										
90	mc	2 Yaquina Head	Yaquina Head	OR										
90	me	2 Tillamook Bay	Hobsonville Point	OR		•								
90	me	2 Columbia River	South Jetty	OR										
90	me	1 Columbia River	North Jetty	WA										

Yr	Sp	Main Location	Specific Location	St	As	Cd	Hg	Ni	Se	tPCB	tDDT	tCdne	tPAH	tBT
90	me	1 Willapa Bay	Nahcotta	WA				•	•				•	
90	mc	2 Gray's Harbor	Westport Jetty	WA										
90	mc	1 Str. Juan de Fuca	Cape Flattery	WA										
90	me	4 South Puget Sound	Budd Inlet	WA										
90	me	4 Commencement Bay	Tahlequah Point	WA									•	
90	me	5 Puget Sound	South Seattle	WA									•	
90	me	5 Elliott Bay	Four-Mile Rock	WA									•	
90	me	5 Elliott Bay	Duwamish Head	WA									•	
90	me	4 Sinclair Inlet	Waterman Point	WA										
90	me	2 Puget Sound	Hood Canal	WA									•	
90	me	4 Whidbey Island	Possession Point	WA				•						
90	me	4 Puget Sound	Everett Harbor	WA									•	
90	me	2 Puget Sound	Port Townsend	WA						•			•	
90	me	2 Puget Sound	Port Angeles	WA										
90	me	3 Bellingham Bay	Squalicum Mar.Jetty	WA				•					•	•
90	me	1 Point Roberts	Point Roberts	WA				•						
90	me	1 Unakwit Inlet	Siwash Bay	AK					•					
90	me	1 Port Valdez	Mineral Creek Flats	AK				•						
90	os	4 Barber's Point	Boat Basin	HI	•			•						•
90	os	5 Honolulu Harbor	Keehi Lagoon	HI	•		•					•	•	•
88	os	2 Kauai	Nawiliwili Harbor	HI				•	•					

Appendix B

This appendix lists the 141 sites sampled in at least 4 of the first 5 years of the NS&T Mussel Watch Project. The number of years is indicated in the column headed "yr." Increasing (I) or decreasing (D) trends under the headings of chemical symbols indicate site/chemical combinations where the Spearman rank correlation coefficient between concentration and year is ≥ 0.9 (I) or ≤ -0.9 (D).

Appendix B. Trends at Mussel Watch Sites.

0 Site	y r	Main Location	Specific Location	St	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn	tPCB	tDDT	tCdne	tPAH
		NORTH ATLANTIC																
4 PBSI	5	Penobscot Bay	Sears Island	ME	-	-	-	-	-	-	-	-	-	-	-	-	-	-
5 PBPI	5	Penobscot Bay	Pickering Island	ME	-	-	I	-	-	-	-	-	-	-	-	-	-	-
9 CAGH	4	Cape Ann	Gap Head	MA	-	I	-	-	-	-	-	-	-	-	D	-	-	-
12 BHDI	5	Boston Harbor	Deer Island	MA	-	-	-	-	-	-	-	-	-	-	-	-	-	-
13 BHDB	5	Boston Harbor	Dorchester Bay	MA	-	-	D	-	-	-	-	-	-	-	-	-	-	-
14 BHDB	5	Boston Harbor	Hingham Bay	MA	-	-	-	-	-	-	-	-	-	-	-	-	-	-
15 BHBI	5	Boston Harbor	Brewster Island	MA	-	-	I	-	-	-	-	-	D	-	-	-	D	D
		MID-ATLANTIC																
17 BBRH	5	Buzzards Bay	Round Hill	MA	-	I	-	-	-	-	-	-	-	-	-	-	-	-
18 BBAR	5	Buzzards Bay	Angelica Rock	MA	I	-	-	-	-	-	-	-	-	-	-	-	-	-
19 BBGN	5	Buzzards Bay	Goosebury Neck	MA	-	-	-	-	-	-	-	-	D	-	-	-	-	-
22 NBBI	5	Narragansett Bay	Dyer Island	RI	-	-	-	-	D	-	-	D	-	-	-	-	-	-
25 BIBI	4	Block Island	Block Island	RI	-	I	-	-	D	-	-	-	-	-	D	-	-	-
27 LICR	5	Long Island Sound	Connecticut River	CT	-	-	D	-	-	I	-	-	-	-	-	-	-	-
28 LINH	5	Long Island Sound	New Haven	CT	D	-	D	-	-	-	-	D	-	-	-	-	-	-
29 LIHR	5	Long Island Sound	Housatonic River	CT	-	-	-	-	-	-	-	-	-	-	-	-	-	-
30 LISI	5	Long Island Sound	Sheffield Island	CT	-	-	-	-	D	-	-	-	-	-	-	-	-	-
32 LIHU	5	Long Island Sound	Huntington Harbor	NY	-	-	-	I	D	-	-	-	D	-	-	-	-	-
33 LIPJ	5	Long Island Sound	Port Jefferson	NY	D	-	-	-	D	-	-	-	-	-	-	-	-	-
34 LIHR	5	Long Island Sound	Mamaroneck	NY	D	D	D	D	D	I	-	-	-	D	-	-	-	-
35 LIHH	5	Long Island Sound	Hempstead Harbor	NY	-	D	D	D	D	-	-	-	-	D	-	-	-	-
36 LITN	5	Long Island Sound	Throgs Neck	NY	D	-	D	-	D	-	-	-	-	D	-	-	-	-
37 MBTH	5	Moriches Bay	Tuthill Point	NY	-	-	-	-	-	-	-	-	-	I	-	-	-	-
37 HRJB	5	Hud./Rar. Estuary	Jamaica Bay	NY	-	-	-	-	-	-	-	-	-	-	-	-	-	-
38 HRUB	5	Hud./Rar. Estuary	Upper Bay	NY	D	-	-	-	-	-	-	-	-	-	I	-	-	-
39 HRLB	5	Hud./Rar. Estuary	Lower Bay	NY	-	-	-	-	-	-	-	-	-	-	-	-	-	-
42 NYSH	5	New York Bight	Sandy Hook	NJ	D	I	-	-	-	-	-	-	-	-	-	-	-	-
43 NYLB	5	New York Bight	Long Branch	NJ	D	-	-	-	-	I	-	-	-	-	-	-	-	-
44 NYSR	5	New York Bight	Shark River	NJ	-	-	D	-	-	-	-	-	-	-	-	D	-	-
49 DBFE	4	Delaware Bay	False Egg Island Point	NJ	D	-	-	-	-	I	-	-	-	-	-	-	-	-
50 DBBD	5	Delaware Bay	Ben Davis Point Shoal	NJ	D	-	-	-	-	-	-	-	-	-	-	-	-	-
51 DBAP	4	Delaware Bay	Arnolds Point Shoal	NJ	D	D	-	-	-	-	I	-	-	-	D	-	-	-

0 Site	y r Main Location	Specific Location	St.	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn	tPCB	DDT	tCd	ne tPAH
52 DBKI	5 Delaware Bay	Kelly Island	DE	D	D	-	-	-	-	-	-	-	-	-	-	-	-
54 CBMP	5 Chesapeake Bay	Mountain Point Bar	MD	-	D	-	-	-	-	-	-	-	-	-	-	-	-
55 CBHP	5 Chesapeake Bay	Hackett Point Bar	MD	-	-	-	-	-	I	I	-	-	-	-	-	-	D
56 CBHG	5 Chesapeake Bay	Hog Point	MD	-	-	-	-	-	-	-	-	-	-	D	D	-	-
59 CBCC	5 Chesapeake Bay	Cape Charles	VA	-	D	-	-	-	-	-	-	-	-	-	-	D	D
61 CBDP	5 Chesapeake Bay	Dandy Point	VA	-	-	-	-	-	-	-	-	-	-	-	-	-	-
63 CBCI	5 Chincoteague Bay	Chincoteague Inlet	VA	-	-	-	-	-	I	D	-	-	-	-	-	-	-
64 QUIB	5 Quinby Inlet	Upshur Bay	VA	-	-	-	-	D	-	-	I	-	D	-	-	-	D
SOUTHEAST ATLANTIC																	
65 RSJC	5 Roanoke Sound	John Creek	NC	-	-	-	-	-	-	-	-	-	-	-	-	-	D
66 PSWB	5 Pamlico Sound	Wysocking Bay	NC	-	-	-	-	I	-	-	-	-	-	-	-	-	-
68 CFB	5 Cape Fear	Battery Island	NC	-	-	-	-	-	-	D	-	-	-	-	-	-	-
69 CHFJ	5 Charleston Harbor	Fort Johnson	SC	-	-	-	-	-	-	-	-	-	-	-	-	-	-
70 CHSF	5 Charleston Harbor	Shutes Folly Island	SC	-	-	-	-	-	-	-	-	-	-	D	-	-	-
72 SRTI	5 Savannah R. Estuary	Tybee Island	GA	-	-	-	-	-	-	-	-	-	-	-	D	-	-
73 SSSI	5 Sapelo Sound	Sapelo Island	GA	-	-	-	-	-	-	-	-	-	-	-	-	-	-
75 SUCB	5 St. Johns River	Chicopit Bay	FL	-	-	-	-	I	-	-	-	-	-	-	-	-	-
77 MRCB	5 Matanzas River	Crescent Beach	FL	-	-	-	-	-	-	-	-	-	-	-	D	-	-
GULF OF MEXICO																	
81 EVFU	5 Everglades	Faka Union Bay	FL	-	-	-	-	-	-	-	I	-	-	-	I	-	-
82 RBHC	5 Rookery Bay	Henderson Creek	FL	-	-	-	-	-	-	-	-	-	-	-	-	-	-
83 NBNB	5 Naples Bay	Naples Bay	FL	-	-	-	-	-	-	-	-	-	-	-	-	-	D
84 CBBI	5 Charlotte Harbor	Bird Island	FL	-	-	-	-	-	-	-	-	-	-	-	-	-	-
88 TBMK	5 Tampa Bay	Mullet Key Bayou	FL	-	D	-	-	-	-	-	-	-	-	-	-	-	-
89 TBCB	5 Tampa Bay	Cockroach Bay	FL	-	-	-	-	-	-	-	I	-	-	-	-	-	-
91 TBPB	5 Tampa Bay	Papys Bayou	FL	-	-	-	-	-	-	-	-	-	-	-	-	-	-
92 CKBP	5 Cedar Key	Black Point	FL	I	-	-	-	-	-	-	-	-	-	-	D	-	-
94 APCP	5 Apalachicola Bay	Cat Point Bar	FL	-	-	-	-	-	-	-	-	-	-	-	-	-	-
95 APDB	5 Apalachicola Bay	Dry Bar	FL	-	-	-	-	-	-	-	-	-	-	-	-	-	-
98 SAWB	5 St. Andrew Bay	Watson Bayou	FL	-	-	-	-	-	-	-	-	-	-	-	-	-	D
99 CBR	5 Choctawhatchee Bay	Off Santa Rosa	FL	-	-	-	I	-	-	-	-	-	-	-	-	-	-
100 CBPP	5 Choctawhatchee Bay	Postil Point	FL	-	-	I	-	-	-	-	-	-	-	-	-	-	-
103 PBIB	4 Pensacola Bay	Indian Bayou	FL	-	-	D	-	D	-	I	-	-	-	-	-	-	D
105 MBGP	5 Mobile Bay	Cedar Point Reef	AL	D	-	-	-	-	-	-	-	-	-	-	-	-	D

0 Site	yr	Main Location	Specific Location	St.	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn	tPCB	tDDT	tCdne	tPAH
109 MSPB	5	Mississippi Sound	Pascagoula Bay	MS	-	-	-	D	-	-	-	-	-	-	-	-	-	-
110 MSBB	5	Mississippi Sound	Biloxi Bay	MS	-	-	-	D	-	-	D	-	-	-	-	-	D	-
111 MSPC	5	Mississippi Sound	Pass Christian	MS	-	-	-	-	I	-	-	I	-	-	-	-	-	-
115 LBMP	5	Lake Borgne	Maheureux Point	LA	-	-	-	-	-	-	-	-	-	-	-	-	-	-
116 BSBG	5	Breton Sound	Bay Gardene	LA	-	-	-	-	-	-	-	-	-	-	-	-	-	-
117 BSSI	5	Breton Sound	Sable Island	LA	-	-	-	-	-	-	-	-	-	-	-	-	-	-
120 BBSD	5	Barataria Bay	Bayou Saint Denis	LA	-	-	-	-	I	-	-	I	-	-	-	-	-	-
122 BBMB	5	Barataria Bay	Middle Bank	LA	-	-	-	-	-	-	-	I	-	-	-	-	D	-
124 TBLF	5	Terrebonne Bay	Lake Felicity	LA	-	D	-	-	-	-	D	-	-	-	-	-	-	-
125 TBLB	5	Terrebonne Bay	Lake Barre	LA	-	D	-	I	-	I	-	-	-	I	-	-	-	-
126 CLCL	5	Caillou Lake	Caillou Lake	LA	-	-	-	-	-	-	-	-	-	-	-	-	-	-
127 ABOB	5	Atchafalaya Bay	Oyster Bayou	LA	-	-	-	-	-	-	-	-	-	-	-	-	-	-
128 VBSP	5	Vermillion Bay	Southwest Pass	LA	-	-	-	-	-	-	-	-	-	-	I	-	-	-
129 JHJH	5	Joel Harbor Bayou	Joseph Harbor Bay	LA	-	-	-	-	I	-	-	-	I	-	-	D	-	-
131 CLSJ	5	Calcasieu Lake	St. Johns Island	LA	-	-	-	-	I	-	-	-	-	-	-	-	D	-
132 SLBB	5	Sabine Lake	Blue Buck Point	TX	-	-	-	-	-	-	-	-	-	D	-	-	-	-
134 GHR	5	Galveston Bay	Hanna Reef	TX	-	-	-	-	I	-	-	-	-	-	-	-	-	-
136 GBYC	5	Galveston Bay	Yacht Club	TX	-	-	-	-	-	-	-	-	-	-	D	D	-	-
137 GBTD	5	Galveston Bay	Todd's Dump	TX	I	-	-	-	I	-	D	-	-	-	-	-	-	-
138 GBCR	5	Galveston Bay	Confederate Reef	TX	-	-	-	-	-	-	-	-	-	-	-	I	-	-
142 MBEM	5	Matagorda Bay	East Matagorda	TX	-	-	-	-	-	-	-	-	-	-	-	-	-	-
145 MBTP	5	Matagorda Bay	Tres Palacios Bay	TX	-	-	-	-	I	-	-	-	-	I	D	-	-	-
146 MBGP	4	Matagorda Bay	Gallinipper Point	TX	-	-	-	-	-	I	-	I	-	-	-	-	-	I
147 MBLR	5	Matagorda Bay	Lavaca River Mouth	TX	-	-	-	-	-	-	-	-	-	-	-	-	-	-
153 MBAR	5	Mesquite Bay	Ayres Reef	TX	I	I	I	-	-	-	-	-	-	I	-	-	-	-
154 OBCR	5	Copano Bay	Copano Reef	TX	-	D	-	-	-	-	-	-	-	-	-	-	-	-
156 ABLR	5	Aransas Bay	Long Reef	TX	D	-	-	-	-	-	-	-	-	-	-	-	-	-
158 OCJC	4	Corpus Christi	Ingliside Cove	TX	-	-	-	-	-	-	-	-	-	I	D	D	-	D
159 OCNB	5	Corpus Christi	Nueces Bay	TX	-	-	-	-	-	-	D	-	-	-	-	-	D	-
161 LMBS	5	L. Laguna Madre	South Bay	TX	-	D	D	-	-	-	D	-	-	-	-	-	-	I

0	Site	y r	Main Location	Specific Location	St.	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn	tPCB	DDT	tCd	ne	tPAH
164	IBNJ	5	WEST COAST	North Jetty	CA	-	-	-	-	-	D	-	-	-	-	-	-	-	-	-
166	SDHI	5	Imperial Beach	Harbor Island	CA	-	-	-	-	-	-	D	D	D	-	-	-	-	-	-
168	PLH	5	Point Loma	Lighthouse	CA	-	I	-	I	-	-	-	-	-	-	D	D	D	-	-
169	MBVB	5	Mission Bay	Ventura Bridge	CA	-	-	-	-	-	-	D	D	-	-	-	-	-	-	D
170	LJLJ	5	La Jolla	Point La Jolla	CA	-	-	-	-	-	-	-	-	D	-	-	-	-	-	-
171	OSBJ	5	Oceanside	Municipal Beach Jetty	CA	-	-	D	-	-	-	-	-	-	-	-	-	-	-	-
173	NBWJ	5	Newport Beach	Wedge Jetty	CA	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
174	ABWJ	5	Anaheim Bay	West Jetty	CA	-	-	-	-	-	-	-	D	-	D	-	-	-	-	-
179	SPFP	5	San Pedro Harbor	Fishing Pier	CA	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
180	PVRP	5	Palos Verdes	Royal Palms State Park	CA	-	-	I	-	-	-	-	-	-	I	-	-	-	-	-
181	SOBR	4	Santa Catalina Is.	Bird Rock	CA	-	-	-	-	-	-	-	-	-	-	-	D	-	-	I
183	MDSJ	5	Marina Del Rey	South Jetty	CA	-	-	D	-	-	-	-	D	-	-	-	-	-	-	-
184	PDPD	5	Point Dume	Point Dume	CA	-	-	-	-	-	-	-	-	-	-	-	D	-	-	-
185	SOFP	4	Santa Cruz Island	Fraser Point	CA	-	-	-	-	-	-	-	-	-	-	-	D	-	-	I
187	SSSB	5	Point Santa Barbara	Point Santa Barbara	CA	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
188	POPC	5	Point Conception	Point Conception	CA	-	-	-	-	-	-	D	-	-	-	-	-	-	-	-
189	SLSL	5	San Luis Ob. Bay	Point San Luis	CA	-	-	I	-	-	I	-	-	-	-	-	-	-	-	-
190	SSSS	5	San Simeon Point	San Simeon Point	CA	-	-	I	-	-	-	-	-	-	-	-	-	-	-	-
191	PGLP	5	Pacific Grove	Lovers Point	CA	-	-	I	-	-	-	-	-	-	-	-	D	-	D	-
192	MBSC	5	Monterey Bay	Point Santa Cruz	CA	-	-	-	-	-	-	-	-	-	-	-	D	-	-	-
201	SFDB	5	San Francisco Bay	Dumbarton Bridge	CA	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
202	SFSM	5	San Francisco Bay	San Mateo Bridge	CA	-	-	-	-	-	-	-	-	I	I	-	-	-	-	-
203	SFEM	4	San Francisco Bay	Emeryville	CA	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
207	TBSR	5	Tomales Bay	Spenger's Residence	CA	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
208	BBBE	5	Bodega Bay	Bodega Bay Entrance	CA	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
210	PALH	5	Point Arena	Lighthouse	CA	I	-	I	-	-	-	-	-	-	-	-	-	-	-	I
211	PDSC	5	Point Delgada	Shelter Cove	CA	-	-	-	-	I	-	-	-	D	D	-	-	-	-	-
212	HMBJ	5	Humboldt Bay	Jetty	CA	-	-	-	-	-	-	-	D	-	-	-	-	-	-	-
214	SSSG	5	Point St. George	Point St. George	CA	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
216	CECH	5	Coos Bay	Coos Head	OR	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
217	CBRP	5	Coos Bay	Russell Point	OR	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
218	YBOP	5	Yaquina Bay	Oneatta Point	OR	-	-	D	-	-	D	D	D	-	-	-	-	-	-	-

0 Site	y r	Main Location	Specific Location	St.	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn	tPCB	tDDT	tCdne	tPAH
219	YHYH	5 Yaguina Head	Yaguina Head	OR	-	-	-	-	-	-	-	-	-	-	-	-	-	-
221	TBHP	5 Tillamook Bay	Hobsonville Point	OR	-	-	-	-	-	-	D	-	-	-	-	-	-	-
222	CRSJ	5 Columbia River	South Jetty	OR	I	-	-	-	-	-	-	-	-	-	-	-	-	-
225	GHMJ	5 Gray's Harbor	Westport Jetty	WA	I	-	-	-	-	-	I	-	-	-	-	-	-	-
226	JCF	4 Str. Juan de Fuca	Cape Flattery	WA	I	-	-	-	-	-	-	-	-	-	-	-	-	-
228	SSBI	5 South Puget Sound	Budd Inlet	WA	-	-	-	-	-	-	-	-	-	-	-	-	-	D
231	CBTP	5 Commencement Bay	Tahlequah Point	WA	-	-	-	-	D	-	-	-	-	-	-	-	-	-
233	EBFR	5 Elliott Bay	Four-Mile Rock	WA	-	-	-	-	D	D	-	-	-	-	D	-	-	-
235	SWP	5 Sinclair Inlet	Waterman Point	WA	-	-	-	-	-	-	-	-	-	-	-	-	-	-
236	WIPP	5 Whidbey Island	Possession Point	WA	-	-	-	-	-	D	-	-	-	-	-	-	-	-
237	BBSM	5 Bellingham Bay	Squalicum Marina Jetty	WA	I	-	-	-	-	-	-	-	-	I	-	-	-	-
238	PRPR	5 Point Roberts	Point Roberts	WA	-	-	-	-	-	-	-	-	-	-	-	-	-	-
241	USB	4 Unakwit Inlet	Siwash Bay	AK	-	-	-	-	-	-	-	-	-	-	-	-	-	-
242	PVMC	4 Port Valdez	Mineral Creek Flats	AK	-	-	-	-	-	-	-	-	D	-	-	-	D	-
245	BPBP	4 Barber's Point	Boat Basin	HI	-	-	-	-	-	D	-	-	D	-	-	-	-	-
246	HHKL	4 Honolulu Harbor	Keehi Lagoon	HI	-	-	D	-	-	-	I	-	D	-	-	-	-	-